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INVENTOR(S)

Yukio NISHIMURA et al.

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TITLE OF APPLICATION

RADIATION-SENSITIVE RESIN COMPOSITION

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Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Dear Sir:

Applicant's hereby submit a Certified English translation and verification of
Japanese Application No: 2002-315021 filed on October 29, 2002.

Respectfully submitted,

MERCHANT & GOULD, P.C.

Christopher W. Raimund
Registration No. 47,258

April 25, 2007

Date

P.O. Box 2903
Minneapolis, Minnesota 55402-0903
Telephone No. (202) 326-0300
Facsimile No. (202) 326-0778

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Patent & Trademark Office



VERIFICATION

I, Yoshinori HAGINO

Kojima International Patent Office
ATSUTA DAIDOSEIMEI-Bldg.2F,
7-26, Jingu 3-chome, Atsuta-ku,
Nagoya-shi, Aichi 456-0031 Japan

Verify that to the best of my knowledge and belief the following
is a true English translation made by me of Japanese patent
Application No.2002-315021 filed on October 29, 2002.

Dated this 9th day of April 2007

Yoshinori Hagino
Yoshinori HAGINO



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[Inventor]
[Address or Domicile] c/o JSR Corporation
11-24, Tsukiji 2-chome, Chuo-ku, Tokyo
[Name] Yukio Nishimura

[Inventor]
[Address or Domicile] c/o JSR Corporation
11-24, Tsukiji 2-chome, Chuo-ku, Tokyo
[Name] Hiroyuki Ishii

[Inventor]
[Address or Domicile] c/o JSR Corporation
11-24, Tsukiji 2-chome, Chuo-ku, Tokyo
[Name] Isao Nishimura

[Inventor]
[Address or Domicile] c/o JSR Corporation
11-24, Tsukiji 2-chome, Chuo-ku, Tokyo
[Name] Eiichi Kobayashi

[Applicant for Patent]
[Identification No.] 000004178
[Name] JSR Corporation

[Agent]
[Identification No.] 100094190
[Patent Attorney]
[Name] Kojima Seiji
[Telephone No.] 052-682-8361

[Agent appointed]
[Identification No.] 100111752
[Patent Attorney]
[Name] Taniguchi Naoya
[Telephone No.] 052-682-8361

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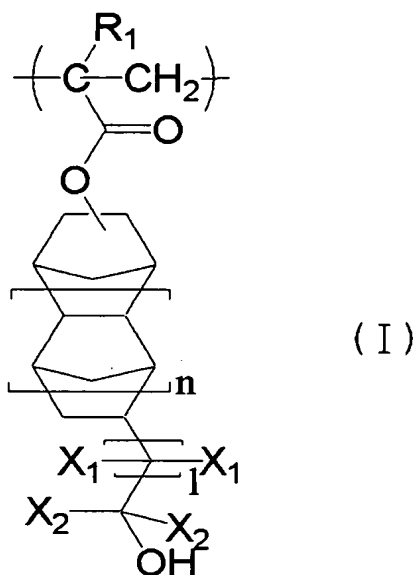
SPECIFICATION

[TITLE OF THE INVENTION]

RADIATION-SENSITIVE RESIN COMPOSITION

[CLAIMS]

[Claim 1] A radiation-sensitive resin composition characterized by comprising [A] a resin which comprises a repeating unit represented by the following formula (I) and is insoluble or scarcely soluble in alkali but becomes alkali soluble by the action of an acid and [B] a radiation-sensitive acid generator.



(In the formula, R₁ represents hydrogen atom, methyl group, a hydroxyalkyl group having 1-4 carbon atoms, or a perfluoroalkyl group having 1-4 carbon atoms, X₁ and X₂ individually represent hydrogen atom, fluorine atom, an alkyl group having 1-4 carbon atoms, or a fluoroalkyl group having 1-4 carbon atoms, l is an integer of 0-5, and n is an integer of 0-2.)

[Claim 2] The radiation-sensitive resin composition according to Claim 1, further comprising [C] an acid diffusion controller.

[DETAILED DESCRIPTION OF THE INVENTION]

[0001]

[Technical Field of the Invention]

The present invention relates to a radiation-sensitive resin composition and more particularly, it relates to a radiation-sensitive resin composition which is useful as a chemically amplified resist in microfabrication with various radiations including deep ultraviolet rays by KrF excimer laser, ArF excimer laser and the like, charged particle rays such as electron beams, and X-rays such as synchrotron radiation.

[0002]

[Prior Art]

In the field of microfabrication represented by the manufacture of integrated circuit

devices, lithographic technology enabling microfabrication with a line width of 0.20 μm or less has been demanded in order to increase the degree of integration in recent years.

In a conventional lithographic process, near ultraviolet rays such as i-line radiation have been generally used. However, it is difficult to perform microfabrication with a line width of a sub-quarter micron level using near ultraviolet rays.

Therefore, in order to enable microfabrication with a line width of 0.20 μm or less, utilization of radiation with a shorter wavelength has been studied. Deep ultraviolet rays represented by a bright line spectrum of a mercury lamp and an excimer laser, X-rays, electron beams, and the like can be given as radiation with a shorter wavelength. Of these, a KrF excimer laser (wavelength: 248 nm) and an ArF excimer laser (wavelength: 193 nm) have gained attention.

As a radiation-sensitive resin composition applicable to the excimer laser radiation, a number of compositions utilizing a chemical amplification effect between a component having an acid-dissociable functional group and a component generating an acid (hereinafter called "acid generator") upon irradiation (hereinafter called "exposure") have been proposed. Such composition is hereinafter called a chemically-amplified radiation-sensitive composition.

As the chemically-amplified radiation-sensitive composition, Patent Document 1 discloses a composition comprising a polymer containing t-butyl ester group of carboxylic acid or t-butylcarbonate group of phenol and an acid generator. This composition utilizes the effect of the polymer to release t-butyl ester group or t-butyl carbonate group in the polymer by the action of an acid generated upon exposure to form an acidic group such as carboxylic group and phenolic hydroxyl group, which allows the exposed area on the resist film to be readily soluble in an alkaline developer.

[0003]

Many conventional chemically-amplified radiation-sensitive compositions use a phenol resin as a base. If the composition contains such a resin, deep ultraviolet rays used as radiation for exposure are absorbed in the resin due to aromatic rings and cannot sufficiently reach the lower layers of the resist film. Because of this, the dose of the radiation is greater in the upper layers and is smaller in the lower layers of the resist film. This causes a resist pattern to be thinner in the upper portion but to be thicker toward the lower portion, thereby forming a trapezoid shape after development. No sufficient resolution can be obtained from such a resist film. If the resist pattern after development is in the shape of a trapezoid, desired dimensional accuracy cannot be achieved in a succeeding step such as an etching step or ion implantation step. Moreover, if the shape of the upper part of the resist pattern is not rectangular, the rate of removal of the resist by dry etching is increased, whereby it is difficult to control etching conditions.

[0004]

On the other hand, the shape of the resist pattern can be improved by increasing the radiation transmittance of the resist film. For example, (meth)acrylate resins represented by polymethylmethacrylate are desirable from the viewpoint of radiation transmittance due to the

superior transparency to deep ultraviolet rays. Patent Document 2 discloses a chemically-amplified radiation-sensitive resin composition using a methacrylate resin. However, this composition has insufficient dry etching resistance due to the absence of an aromatic ring, although the composition excels in microfabrication performance. Therefore, this composition also has difficulty in performing etching with high accuracy and cannot satisfy transparency to radiation and dry etching resistance at the same time.

[0005]

A method of introducing an alicyclic ring into the resin component in the composition instead of an aromatic ring has been known as a means of improving dry etching resistance without impairing radiation transmittance of the resist made from a chemically-amplified radiation-sensitive resin composition. A chemically-amplified radiation-sensitive resin composition using a (meth)acrylate resin having an alicyclic ring is proposed in Patent Document 3, for example.

This composition, however, comprises groups which are comparatively easily dissociated by conventional acids (for example, an acetal functional group such as tetrahydropyranyl group) and groups which are comparatively difficult to be dissociated by an acid (for example, a t-butyl-based functional group such as t-butyl ester group and t-butylcarbonate group) as an acid-dissociable functional group in the resin component. The resin component having the former acid-dissociable functional group provides a resist with favorable basic properties, particularly excellent sensitivity and pattern formability, but the composition has a problem with storage stability. The resin component having the latter acid-dissociable functional group, on the other hand, provides a composition with good storage stability, but the resist has a problem with its basic properties, particularly with sensitivity and pattern formability. In addition, inclusion of an alicyclic structure in the resin components of this composition increases hydrophobicity of the resin, resulting in poor adhesion to substrates.

In view of recent progress in the microfabrication of semiconductor devices, development of a novel resin component exhibiting high transmittance of radiations, having excellent basic properties as a resist, and suitable for use in chemically-amplified radiation-sensitive compositions which can be adapted for short wavelength radiations represented by deep ultraviolet rays has been undertaken (for example, refer to Patent Document 4). However, there have been no resin components satisfying all of these requirements.

[0006]

[Patent Document 1] JP-B-H02-27660

[Patent Document 2] JP-A-H04-226461

[Patent Document 3] JP-A-H07- 234511

[Patent Document 4] JP-A-2002-72484

[0007]

[Problems to be solved by the Invention]

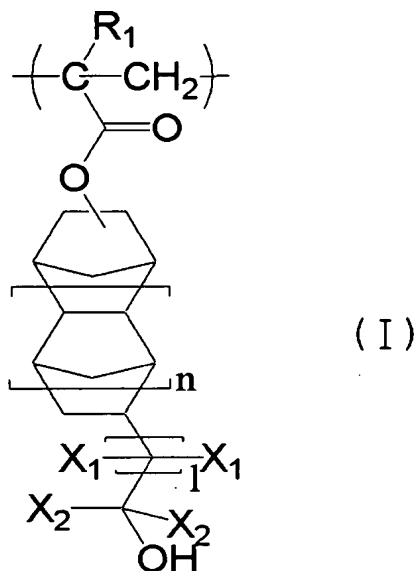
An objective of the present invention is to provide a radiation-sensitive resin

[0008]
[Means for solving problem]

The radiation-sensitive resin composition of the present invention is characterized by comprising [A] a resin which comprises a repeating unit represented by the following formula (I) and is insoluble or scarcely soluble in alkali but becomes alkali soluble by the action of an acid and [B] a radiation-sensitive acid generator.

[Means for solving problem]

The radiation-sensitive resin composition of the present invention is characterized by comprising [A] a resin which comprises a repeating unit represented by the following formula (I) and is insoluble or scarcely soluble in alkali but becomes alkali soluble by the action of an acid and [B] a radiation-sensitive acid generator.

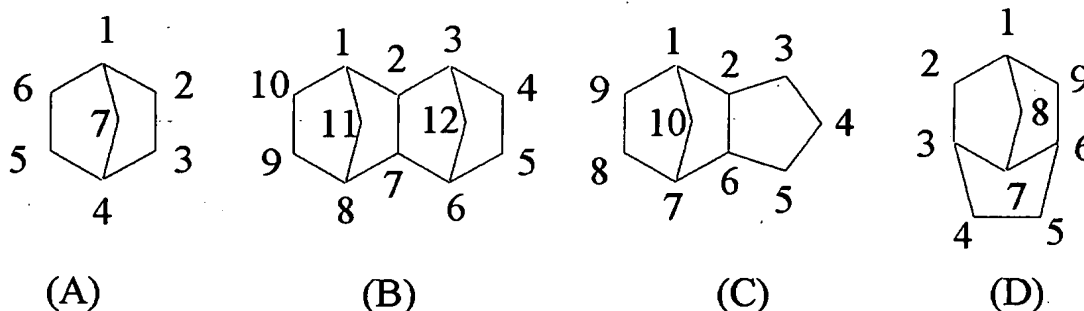


[0009]

[0010]

The present invention will now be described in detail.

4



In the above formulas, the skeleton (A) is named bicyclo[2.2.1]heptane, (B) is named tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecane, (C) is named tricyclo[5.2.1.0^{2,6}]decane, and (D) is named tricyclo[4.2.1.0^{3,7}]nonane. In the following description, the nomenclatures (A) to (D) are followed.

[0011]

1. Resin [A]

The resin [A] contained in the radiation-sensitive resin composition of the present invention is a polymer comprising a repeating unit represented by the above formula (I) (hereinafter referred to as “repeating unit (1)”).

The resin [A] of the present invention is insoluble or scarcely soluble in alkali, but becomes easily soluble in alkali by the action of an acid. The term “alkali insoluble or scarcely alkali-soluble” used herein indicates the following properties of the resin [A]. Specifically, in the case of developing a film using only the resin [A] instead of a resist film under alkaline development conditions employed when forming a resist pattern of the resist film formed from a radiation-sensitive resin composition containing the resin [A], the term “alkali insoluble or scarcely alkali-soluble” refers to properties in which 50% or more of the initial thickness of the resist film remains after the development.

[0012]

The substituent R₁ constituting the above repeating unit (1) is one selected from hydrogen atom, methyl group, a hydroxyalkyl group having 1-4 carbon atoms, and a perfluoroalkyl group having 1-4 carbon atoms. Examples are hydrogen atom, methyl group, ethyl group, monofluoromethyl group, difluoromethyl group, trifluoromethyl group, hydroxymethyl group, 2-hydroxyethyl group, and the like. Of these, hydrogen atom, methyl group, trifluoromethyl group and hydroxymethyl group are preferable.

[0013]

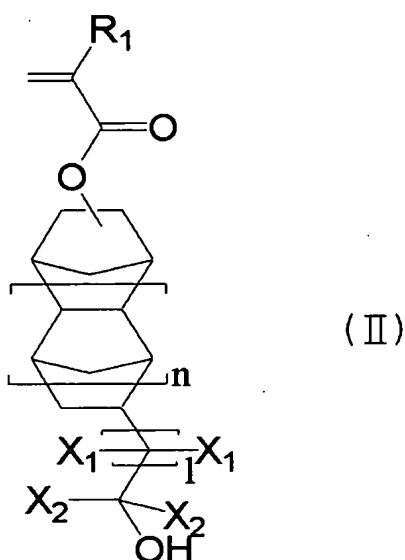
X₁ and X₂ constituting the above repeating unit (1) are individually one selected from hydrogen atom, fluorine atom, an alkyl group having 1-4 carbon atoms, and a fluoroalkyl group having 1-4 carbon atoms. Examples of the alkyl group include methyl group, ethyl group, n-propyl group, i-propyl group, n-butyl group, 2-methylpropyl group, 1-methylpropyl group, t-butyl group and the like. Additionally, examples of the fluoroalkyl group include monofluoromethyl group, difluoromethyl group, trifluoromethyl group, 1-fluoroethyl group,

1,2-difluoroethyl group, 1,1,2,2-tetrafluoroethyl group and the like. The X_1 is preferably hydrogen atom, fluorine atom, methyl group and trifluoromethyl group. The X_2 is preferably fluorine atom and trifluoromethyl group. Further, the preferable combination of X_1 and X_2 includes a combination of hydrogen atom for X_1 and trifluoromethyl group for X_2 , a combination of fluorine atom for X_1 and trifluoromethyl group for X_2 , and the like.

In the formula (I), l is an integer of 0-5, and preferably an integer of 0-3. In addition, n is an integer of 0-2, and preferably 0 or 1.

[0014]

Example of a monomer forming the repeating unit (1) having the above substituents R_1 , X_1 and X_2 includes a compound represented by the following formula (II).



(In the formula, R_1 represents hydrogen atom, methyl group, a hydroxyalkyl group having 1-4 carbon atoms, or a perfluoroalkyl group having 1-4 carbon atoms, X_1 and X_2 individually represent hydrogen atom, fluorine atom, an alkyl group having 1-4 carbon atoms, or a fluoroalkyl group having 1-4 carbon atoms, l is an integer of 0-5, and n is an integer of 0-2.)

[0015]

The compounds represented by the above formula (II) can be obtained by the following method, for example.

1) 5-(2,2-ditrifluoromethyl-2-hydroxyethyl)bicyclo[2.2.1]hept-2(3)-yl formate is obtained by the Michael addition reaction of 5-(2,2-ditrifluoromethyl-2-hydroxyethyl)bicyclo[2.2.1]hept-2-ene with formic acid. ("2(3)-" here indicates that bonding site is 2-position or 3-position and that the compound includes isomers. Hereinafter the same.)

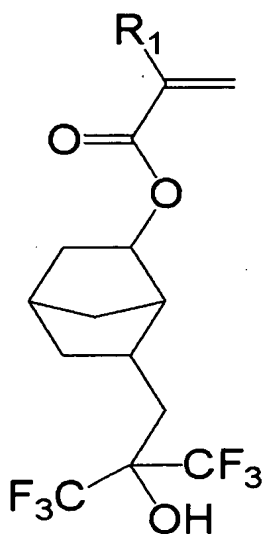
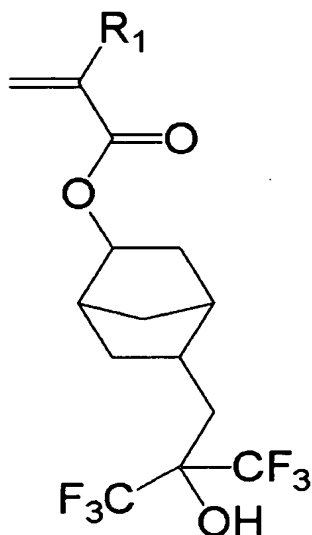
2) The compound obtained in 1) is hydrolyzed with a base such as sodium carbonate to obtain 5-(2,2-ditrifluoromethyl-2-hydroxyethyl)bicyclo[2.2.1]heptan-2(3)-ol.

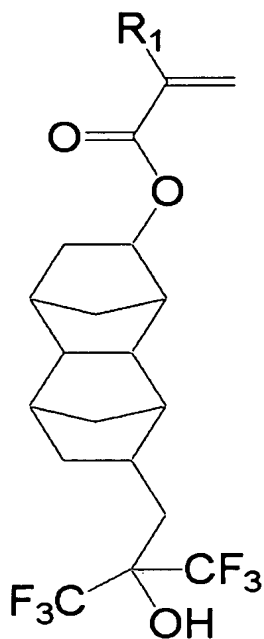
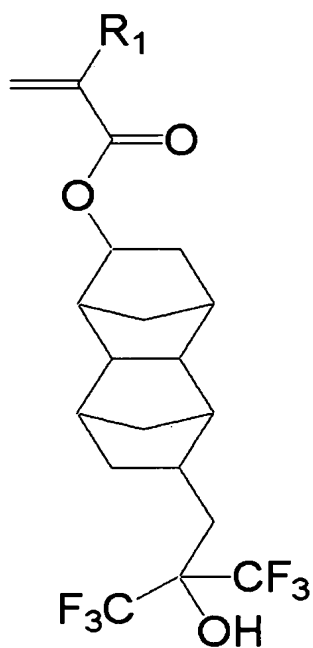
3) Next, the compound obtained in 2) is reacted with, for example, n -butyl lithium in an amount of two equivalents to obtain a compound in which two alcohol are capped,

followed by de-hydrochloric acid reaction in the presence of one equivalent of acid chloride of (meth)acrylic acid and a base to selectively react alcohols at 2-position and/or 3-position with (meth)acrylic acid, thereby obtaining the compound providing the repeating unit (1).

[0016]

The following compounds are particularly preferable among the compounds forming the repeating unit (1).





[0017]

Either one type or two more types of the repeating unit (1) may be present in the resin [A].

[0018]

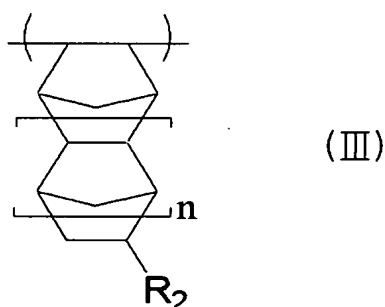
The resin [A] may be of only the repeating unit (1) or of the repeating unit (1) and other repeating units, but is preferably a copolymer comprising the other repeating units. In the latter case, there are no specific limitations to the types and the like of the other repeating

units. Additionally, the content of the repeating unit (1) is usually 10-80 mol%, preferably 10-60 mol%, and further preferably 10-50 mol% based on the total amount of the repeating units. If the content of the repeating unit (1) is less than 10 mol%, solubility in solvents, adhesion to substrates, and developability of the resulting resist tend to decrease. On the other hand, when the content exceeds 80 mol%, resist pattern resolution tends to decrease.

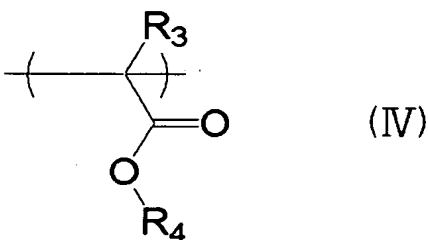
[0019]

Examples of the other repeating units include repeating units represented by the following formulas (III) and (IV).

[0020]



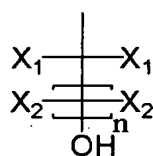
(In the formula, R_2 represents a monovalent organic group and n is an integer of 0-2.)



(In the formula, R_3 represents hydrogen atom, methyl group, a hydroxylalkyl group having 1-4 carbon atoms, or a perfluoroalkyl group having 1-4 carbon atoms and R_4 represents a monovalent organic group.)

[0021]

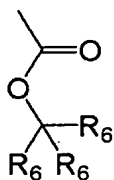
The monovalent organic group R_2 constituting the repeating unit represented by the general formula (III) (hereinafter referred to as "repeating unit (2)") is not particularly limited, but includes organic groups represented by the following formulas (2-1) to (2-4).



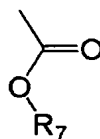
(2-1)



(2-2)



(2-3)



(2-4)

(In the formula (2-1), X_1 and X_2 individually represent hydrogen atom, fluorine atom, an alkyl group having 1-4 carbon atoms, or a fluoroalkyl group having 1-4 carbon atoms, and n is an integer of 0-5. In the organic group (2-2), R_5 may represent a divalent organic group having a linear, branched or cyclic skeleton, in which case, X represents hydrogen atom, hydroxyl group, carboxyl group, nitro group, cyano group, or amino group, or R_5 may not be present, in which case, X is carboxyl group or cyano group. In the organic group (2-3), R_6 individually represents a monovalent alicyclic hydrocarbon group having 4-20 carbon atoms or a derivative thereof, or an alkyl group having 1-4 carbon atoms or a derivative thereof. In the organic group (2-4), R_7 represents an alkyl group having 1-6 carbon atoms or a derivative thereof, a cycloalkyl group having 5-10 carbon atoms or a derivative thereof, a monovalent polyalicyclic hydrocarbon group having 4-20 carbon atoms or a derivative thereof, or a lactone ring.)

[0022]

In the above repeating unit (2), in the case the substituent R_2 is the above organic group (2-1), examples of an alkyl group when X_1 and X_2 are the alkyl group include methyl group, ethyl group, n-propyl group, i-propyl group, n-butyl group, 2-methylpropyl group, 1-methylpropyl group, t-butyl group and the like. Examples of the fluoroalkyl group when X_1 and X_2 are the fluoroalkyl group include monofluoromethyl group, difluoromethyl group, trifluoromethyl group, 1-fluoroethyl group, 1,2-difluoroethyl group, 1,1,2,2-tetrafluoroethyl group and the like. The above X_1 is preferably hydrogen atom, fluorine atom and trifluoromethyl group. The above X_2 is preferably hydrogen atom, fluorine atom and trifluoromethyl group. Therefore, as the organic group (2-1) bonded to the main chain at the 1-position, hydroxymethyl group, 2-hydroxymethyl group, 3-hydroxypropyl group, 1-fluoro-1-hydroxymethyl group, 1,1-difluoro-1-hydroxymethyl group, 1,2-difluoro-2-hydroxymethyl group, 1,1,2,2-tetrafluoro-2-hydroxymethyl group, 2-trifluoromethyl-2-hydroxyethyl group, and 2,2-ditrifluoromethyl-2-hydroxyethyl group are preferable.

[0023]

In the above repeating unit (2), in the case the substituent R_2 is the above organic group (2-2), examples of R_5 constituting the substituent R_2 include an alkylene group such as methylene group, ethylene group, n-propylene group, isopropylene group, n-butylene group and isobutylene group; a divalent organic group derived from a cycloalkane such as cyclopentane, cyclohexane, cycloheptane and cyclooctane; a divalent organic group derived from adamantane; a divalent organic group derived from bicyclo[2.2.1]heptane; a divalent

organic group derived from tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecane, and the like. Of these groups, methylene group, ethylene group, divalent organic group derived from adamantane, and divalent organic group derived from bicyclo[2.2.1]heptane are preferable.

In the above case, X constituting the organic group (2-2) may be hydrogen atom, hydroxyl group, carboxyl group, nitro group, cyano group and amino group. Of these, hydrogen atom, hydroxyl group and nitro group are preferable.

When R₅ is not present in the above organic group (2-2), X may be the substituent R₂ as is. Examples include carboxyl group and cyano group.

[0024]

Therefore, the above organic group (2-2), indicated as a residue bonded to oxygen atom, may be hydrogen atom, hydroxymethyl group, 2-hydroxyethyl group, 3-hydroxypropyl group, 3-hydroxyadamant-1-yl group, 5(6)-hydroxybicyclo[2.2.1]hept-2-yl group, 9(10)-hydroxytetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-yl group, carboxyl group, carboxymethyl group, 2-carboxyethyl group, 3-carboxypropyl group, 3-carboxyadamantan-1-yl group, 5(6)-carboxybicyclo[2.2.1]hept-2-yl group, 9(10)-carboxytetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-yl group, cyano group, cyanomethyl group, 2-cyanoethyl group, 3-cyanopropyl group, 3-cyanoadamantan-1-yl group, 5(6)-cyanobicyclo[2.2.1]hept-2-yl group, 9(10)-cyanotetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-yl group and the like.

[0025]

In the above repeating unit (2), in the case the substituent R₂ is the above organic group (2-3), examples of an alicyclic hydrocarbon group when the substituent R₆ is the alicyclic hydrocarbon group include cyclobutyl group, cyclopentyl group, cyclohexyl group, cycloheptyl group, cyclooctyl group and the like. In addition, examples of the derivative of such alicyclic hydrocarbon group include a group in which hydrogen atom on the alicyclic hydrocarbon group is substituted with one or more substituents such as hydroxyl group; carboxyl group; oxy group (=O); a hydroxyalkyl group having 1-4 carbon atoms such as hydroxymethyl group, 1-hydroxyethyl group, 2-hydroxyethyl group, 1-hydroxypropyl group, 2-hydroxypropyl group, 3-hydroxypropyl group, 2-hydroxybutyl group, 3-hydroxybutyl group, and 4-hydroxybutyl group; an alkoxyl group having 1-4 carbon atoms such as methoxy group, ethoxy group, n-propoxy group, i-propoxy group, n-butoxy group, 2-methylpropoxy group, 1-methylpropoxy group and t-butoxy group; cyano group; a cyanoalkyl group having 2-5 carbon atoms such as cyanomethyl group, 2-cyanoethyl group, 3-cyanopropyl group and 4-cyanobutyl group; and the like. Of these substituents, hydroxyl group, carboxyl group, hydroxymethyl group, cyano group and cyanomethyl group are preferable.

[0026]

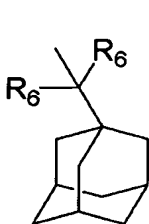
Further, when the substituent R₆ is an alkyl group, examples of the alkyl group include methyl group, ethyl group, n-propyl group, i-propyl group, n-butyl group, 1-methylpropyl group, 2-methylpropyl group, t-butyl group and the like. Of these groups, methyl group and ethyl group are preferable. In addition, examples of the derivative of such alkyl group include a group in which hydrogen atom on the alkyl group is substituted with one or more substituents such as hydroxyl group; carboxyl group; oxy group (=O); a hydroxyalkyl

group having 1-4 carbon atoms such as hydroxymethyl group, 1-hydroxyethyl group, 2-hydroxyethyl group, 1-hydroxypropyl group, 2-hydroxypropyl group, 3-hydroxypropyl group, 2-hydroxybutyl group, 3-hydroxybutyl group and 4-hydroxybutyl group; an alkoxyl group having 1-4 carbon atoms such as methoxy group, ethoxy group, n-propoxy group, i-propoxy group, n-butoxy group, 2-methylpropoxy group, 1-methylpropoxy group and t-butoxy group; cyano group; a cyanoalkyl group having 2-5 carbon atoms such as cyanomethyl group, 2-cyanoethyl group, 3-cyanopropyl group and 4-cyanobutyl group; and the like. Of these substituents, hydroxyl group, carboxyl group, hydroxymethyl group, cyano group, and cyanomethyl group are preferable.

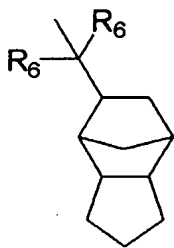
[0027]

Regarding the substituent R_6 in the above organic group (2-3), all three substituents may be the same, two among the three substituents may be the same, or all three substituents may differ from each other.

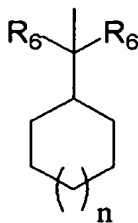
Examples of the organic group (2-3) in which two among three substituents are the same and another substituent R_6 is an alicyclic hydrocarbon group include groups represented below.



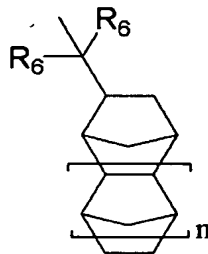
(6-1)



(6-2)



(6-3)

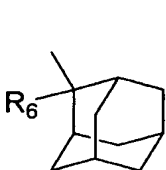


(6-4)

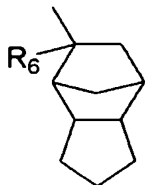
(In the above substituents (6-3) and (6-4), n is an integer of 0-2.)

[0028]

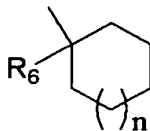
Moreover, the above organic group (2-3) may be a divalent alicyclic hydrocarbon group having 4-20 carbon atoms which is obtained by combining two substituents R_6 , and a derivative thereof. The following examples can be given.



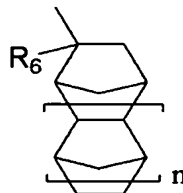
(6-5)



(6-6)



(6-7)



(6-8)

(In the above substituents (6-7) and (6-8), n is an integer of 0-2.)

Examples of the group having the above structure include ones formed by bicyclo[2.2.1]heptane, tricyclo[6.2.1.1.0]^{2,7}decane, tetracyclo[5.2.1.0^{2,6}]dodecane and the like,

and the like.

[0029]

Therefore, as examples of the above organic group (2-3), in terms of the residue bound to oxygen atom in the ester group, which is represented by the group $-C(R_6)_3$, the following groups can be given:

a trialkylmethyl group such as t-butyl group, 3-methyl-3-butyl group, 2-methyl-2-butyl group, 2-ethyl-2-butyl group, 3-ethyl-3-butyl group and 3-methyl-3-pentyl group;

an alkylcycloalkyl group such as 2-methyladamantan-2-yl group, 2-methyl-3-hydroxyadamantan-2-yl group, 2-methyl-3-cyanoadamantan-2-yl group, 2-ethyladamantan-2-yl group, 2-ethyl-3-hydroxyadamantan-2-yl group, 2-ethyl-3-cyanoadamantan-2-yl group, 8-methyltricyclo[5.2.1.0^{2,6}]decan-8-yl group, 8-methyl-4-hydroxytricyclo[5.2.1.0^{2,6}]decan-8-yl group, 8-methyl-4-cyanotricyclo[5.2.1.0^{2,6}]decan-8-yl group, 8-ethyltricyclo[5.2.1.0^{2,6}]decan-8-yl group, 8-ethyl-4-hydroxytricyclo[5.2.1.0^{2,6}]decan-8-yl group, 8-ethyl-4-cyanotricyclo[5.2.1.0^{2,6}]decan-8-yl group, 1-methylcyclopentyl group, 1-ethylcyclopentyl group, 1-methylcyclohexyl group, 1-ethylcyclohexyl group, 2-methylcyclopentyl group, 2-ethylcyclopentyl group, 2-methylbicyclo[2.2.1]hept-2-yl group, 2-methyl-5(6)-hydroxybicyclo[2.2.1]hept-2-yl group, 2-methyl-5-cyanobicyclo[2.2.1]hept-2-yl group, 2-ethylbicyclo[2.2.1]hept-2-yl group, 2-ethyl-5(6)-hydroxybicyclo[2.2.1]hept-2-yl group, 2-ethyl-5(6)-cyanobicyclo[2.2.1]hept-2-yl group, 4-methyltetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl group, 4-methyl-9(10)-hydroxytetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl group, 4-methyl-9(10)-cyanotetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl group, 4-ethyl-tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl group, 4-ethyl-9(10)-hydroxytetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl group and 4-ethyl-9(10)-cyanotetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl group;

a dialkylcycloalkylmethyl group such as 1-cyclopentyl-1-methylethyl group, 1-cyclohexyl-1-methylethyl group, 1-cycloheptyl-1-methylethyl group, 1-bicyclo[2.2.1]hept-2-yl-1-methylethyl group, 1-tricyclo[5.2.1.0^{2,6}]decan-8-yl-1-methylethyl group, 1-tetracyclo[7.5.2.1.0^{2,7}]dodecan-4-yl-1-methylethyl group, 1-adamantan-1-yl-1-methylethyl group, 1-(2(3)-hydroxycyclopentyl)-1-methylethyl group, 1-(3(4)-hydroxycyclohexyl)-1-methylethyl group, 1-(3(4)-hydroxycycloheptyl)-1-methylethyl group, 1-(5(6)-hydroxybicyclo[2.2.1]hept-2-yl)-1-methylethyl group, 1-(4-hydroxytricyclo[5.2.1.0^{2,6}]decan-8-yl)-1-methylethyl group, 1-(9(10)-hydroxytetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl)-1-methylethyl group, 1-(3-hydroxyadamantan-1-yl)-1-methylethyl group, 1-(2(3)-cyanocyclopentyl)-1-methylethyl group, 1-(3(4)-cyanocyclohexyl)-1-methylethyl

group, 1-(3(4)-cyanocycloheptyl)-1-methylethyl group,
1-(5(6)-cyanobicyclo[2.2.1]hept-2-yl)-1-methylethyl group,
1-(4-cyanotricyclo[5.2.1.0^{2,6}]decan-8-yl)-1-methylethyl group,
1-(9(10)-cyanotetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl)-1-methylethyl group and
1-(3-cyanoadamantan-1-yl)-1-methylethyl group;

an alkyldicycloalkylmethyl group such as 1,1-dicyclopentylethyl group,
1,1-dicyclohexylethyl group, 1,1-dicycloheptylethyl group,
1,1-dibicyclo[2.2.1]hept-2-ylethyl group, 1,1-ditricyclo[5.2.1.0^{2,6}]decan-8-ylethyl group,
1,1-di(tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl)ethyl group, 1,1-diadamantan-1-ylethyl group,
1,1-di(2(3)-hydroxycyclopentyl)ethyl group, 1,1-di(3(4)-hydroxycyclohexyl)ethyl group,
1,1-di(3(4)-hydroxycycloheptyl)ethyl group,
1,1-di(5(6)-hydroxybicyclo[2.2.1]hept-2-yl)ethyl group,
1,1-di(4-hydroxytricyclo[5.2.1.0^{2,6}]decan-8-yl)ethyl group,
1,1-di(9(10)-hydroxytetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl)ethyl group,
1,1-di(3-hydroxyadamantan-1-yl)ethyl group, 1,1-di(2(3)-cyanocyclopentyl)ethyl group,
1,1-di(3(4)-cyanocyclohexyl)ethyl group, 1,1-di(3(4)-cyanocycloheptyl)ethyl group,
1,1-di(5(6)-cyanobicyclo[2.2.1]hept-2-yl)ethyl group,
1,1-di(4-cyanotricyclo[5.2.1.0^{2,6}]decan-8-yl)ethyl group,
1,1-di(9(10)-cyanotetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl)ethyl group and
1,1-di(3-cyanoadamantan-1-yl)ethyl group; and the like.

[0030]

Of the above organic groups, t-butyl group, 2-methyl-2-propyl group,
2-methyl-2-butyl group, 2-ethyl-2-butyl group, 3-ethyl-3-butyl group,
2-methyladamantan-2-yl group, 2-methyl-3-hydroxyadamantan-2-yl group,
2-ethyladamantan-2-yl group, 8-methyltricyclo[5.2.1.0^{2,6}]decan-8-yl group,
8-ethyltricyclo[5.2.1.0^{2,6}]decan-8-yl group, 1-methylcyclopentyl group, 1-ethylcyclopentyl
group, 1-methylcyclohexyl group, 1-ethylcyclohexyl group, 2-methylbicyclo[2.2.1]hept-2-yl
group, 2-ethylbicyclo[2.2.1]hept-2-yl group, 4-methyltetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl
group, 4-ethyltetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl group, 1-cyclohexyl-1-methylethyl
group, 1-bicyclo[2.2.1]hept-2-yl-1-methylethyl group,
1-tricyclo[5.2.1.0^{2,6}]decan-8-yl-1-methylethyl group,
1-tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl-1-methylethyl group,
1-adamantan-1-yl-1-methylethyl group, 1-(2(3)-hydroxycyclopentyl)-1-methylethyl group,
1-(3(4)-hydroxycyclohexyl)-1-methylethyl group,
1-(3(4)-hydroxycycloheptyl)-1-methylethyl group,
1-(3-hydroxyadamantan-1-yl)-1-methylethyl group, 1,1-dicyclohexylethyl group,
1,1-dibicyclo[2.2.1]hept-2-ylethyl group, 1,1-ditricyclo[5.2.1.0^{2,6}]decan-8-ylethyl group,
1,1-di(tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl)ethyl group and 1,1-diadamantan-1-ylethyl
group are preferable.

[0031]

In the above repeating unit (2), in the case the substituent R₂ is the above organic

group (2-4), examples of an alkyl group when R₇ constituting the above substituent R₂ is the alkyl group include methyl group, ethyl group, n-propyl group, i-propyl group, n-butyl group, 2-methylpropyl group, 1-methylpropyl group, n-pentyl group, n-hexyl group and the like. Of these groups, methyl group and ethyl group are preferable. In addition, examples of the derivative of such alicyclic hydrocarbon group include a group in which hydrogen atom on the alicyclic hydrocarbon group is substituted with one or more substituents such as hydroxyl group; carboxyl group; oxy group (=O); a hydroxyalkyl group having 1-4 carbon atoms such as hydroxymethyl group, 1-hydroxyethyl group, 2-hydroxyethyl group, 1-hydroxypropyl group, 2-hydroxypropyl group, 3-hydroxypropyl group, 2-hydroxybutyl group, 3-hydroxybutyl group, and 4-hydroxybutyl group; an alkoxyl group having 1-4 carbon atoms such as methoxy group, ethoxy group, n-propoxy group, i-propoxy group, n-butoxy group, 2-methylpropoxy group, 1-methylpropoxy group and t-butoxy group; cyano group; a cyanoalkyl group having 2-5 carbon atoms such as cyanomethyl group, 2-cyanoethyl group, 3-cyanopropyl group and 4-cyanobutyl group; and the like.

[0032]

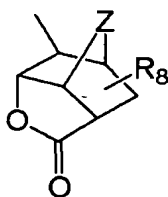
When the substituent R₇ constituting the above substituent R₂ is a cycloalkyl group, examples of the cycloalkyl group include cyclopentyl group, cyclohexyl group, cycloheptyl group, cyclooctyl group and the like. Of these, cyclopentyl group, cyclohexyl group, cycloheptyl group and cyclooctyl group are preferable. In addition, examples of the derivative of such alicyclic hydrocarbon group include a group in which hydrogen atom on the alicyclic hydrocarbon group is substituted with one or more substituents such as hydroxyl group; carboxyl group; oxy group (=O); a hydroxyalkyl group having 1-4 carbon atoms such as hydroxymethyl group, 1-hydroxyethyl group, 2-hydroxyethyl group, 1-hydroxypropyl group, 2-hydroxypropyl group, 3-hydroxypropyl group, 2-hydroxybutyl group, 3-hydroxybutyl group, and 4-hydroxybutyl group; an alkoxyl group having 1-4 carbon atoms such as methoxy group, ethoxy group, n-propoxy group, i-propoxy group, n-butoxy group, 2-methylpropoxy group, 1-methylpropoxy group and t-butoxy group; cyano group; a cyanoalkyl group having 2-5 carbon atoms such as cyanomethyl group, 2-cyanoethyl group, 3-cyanopropyl group and 4-cyanobutyl group; and the like.

[0033]

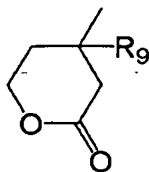
When the substituent R₇ constituting the substituent R₂ is a polyalicyclic hydrocarbon group, examples of the polyalicyclic hydrocarbon group include a group derived from a polyalicyclic hydrocarbon such as adamantane, bicyclo[2.2.1]heptane, 7,7-dimethylbicyclo[2.2.1]heptane, tricyclo[5.2.1.0^{2,6}]decane and tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecane. Of these, groups derived from adamantane, bicyclo[2.2.1]heptane, 7,7-dimethylbicyclo[2.2.1]heptane and tricyclo[5.2.1.0^{2,6}]decane are preferable. In addition, examples of the derivative include a group in which bound hydrogen atom is substituted with one or more functional groups described above.

[0034]

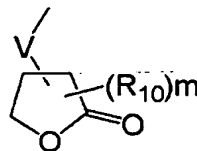
The substituent R₇ constituting the substituent R₂ may also be a group having a lactone ring shown below.



(7-1)



(7-2)



(7-3)

(In the substituent (7-1), R_8 represents hydrogen atom, an alkyl group having 1-5 carbon atoms or an alkoxyl group having 1-5 carbon atoms, and Z represents methylene group, dimethylmethylene group, oxygen atom or sulfur atom. In the substituent (7-2), R_9 represents hydrogen atom, an alkyl group having 1-5 carbon atoms or an alkoxyl group having 1-5 carbon atoms. n is an integer of 0-2. In the substituent (7-3), R_{10} represents hydrogen atom, an alkyl group having 1-5 carbon atoms or an alkoxyl group having 1-5 carbon atoms, and V represents methylene group. Additionally, m is an integer of 0-4.)

[0035]

Examples of the alkyl group used for the substituent R_8 constituting the substituent (7-1), the substituent R_9 constituting the substituent (7-2), and the substituent R_{10} constituting the substituent (7-3) include methyl group, ethyl group, *n*-propyl group, *i*-propyl group, *n*-butyl group, 1-methylpropyl group, 2-methylpropyl group, *t*-butyl group, *n*-pentyl group and the like. In addition, examples of the alkoxyl group used for the substituent R_8 constituting the substituent (7-1), the substituent R_9 constituting the substituent (7-2) and the substituent R_{10} constituting the substituent (7-3) include methoxy group, ethoxy group, *n*-propoxy group, *i*-propoxy group, *n*-butoxy group, 1-methylpropoxy group, 2-methylpropoxy group, *t*-butoxy group, *n*-pentyloxy group and the like.

Z in the above substituent (7-1) may not be present. V in the above substituent (7-3) may not be present.

[0036]

Therefore, as preferable examples of the above organic group (2-4), in terms of the residue bound to oxygen atom in the ester group, the following groups can be given:

an alkyl group such as methyl group, ethyl group, *n*-propyl group, cyclopentyl group, cyclohexyl group, adamantan-1-yl group, bicyclo[2.2.1]hept-2-yl group, 7,7-dimethylbicyclo[2.2.1]hept-1-yl group, tricyclo[5.2.1.0^{2,6}]decan-8-yl group and tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl group;

a group having a lactone ring such as a 5-oxo-4-oxa-tricyclo[4.2.1.0^{3,7}]nonan-2-yl group, 9-methoxycarbonyl-5-oxo-4-oxa-tricyclo[4.2.1.0^{3,7}]nonan-2-yl group, 7-oxo-6-oxabicyclo[3.2.1]octan-4-yl group, 2-methoxycarbonyl-7-oxo-6-oxa-bicyclo[3.2.1]octan-4-yl group, 2-oxotetrahydropyran-4-yl group, 4-methyl-2-oxotetrahydropyran-4-yl group, 4-ethyl-2-oxotetrahydropyran-4-yl group, 4-propyl-2-oxotetrahydropyran-4-yl group, 5-oxotetrahydrofuran-3-yl group, 2,2-dimethyl-5-oxotetrahydrofuran-3-yl group, 4,4-dimethyl-5-oxotetrahydrofuran-3-yl

group, 2-oxotetrahydrofuran-3-yl group, 4,4-dimethyl-2-oxotetrahydrofuran-3-yl group, 5,5-dimethyl-2-oxotetrahydrofuran-3-yl group, 2-oxotetrahydrofuran-3-yl group, 5-oxotetrahydrofuran-2-ylmethyl group, 3,3-dimethyl-5-oxotetrahydrofuran-2-ylmethyl group and 4,4-dimethyl-5-oxotetrahydrofuran-2-ylmethyl group; and the like.

[0037]

Examples of monomer providing the repeating unit (2) are as follows:

5-hydroxymethylbicyclo[2.2.1]hept-2-ene, 5-(2-hydroxyethyl)bicyclo[2.2.1]hept-2-ene, 5-(3-hydroxypropyl)bicyclo[2.2.1]hept-2-ene, 5-(1-fluoro-1-hydroxymethyl)bicyclo[2.2.1]hept-2-ene, 5-(1,1-fluoro-1-hydroxymethyl)bicyclo[2.2.1]hept-2-ene, 5-(1,2-difluoro-2-hydroxymethyl)bicyclo[2.2.1]hept-2-ene, 5-(1,1,2,2-tetrafluoro-2-hydroxymethyl)bicyclo[2.2.1]hept-2-ene, 5-(2-trifluoromethyl-2-hydroxyethyl)bicyclo[2.2.1]hept-2-ene, 5-(2,2-ditrifluoromethyl-2-hydroxyethyl)bicyclo[2.2.1]hept-2-ene,

[0038]

bicyclo[2.2.1]hept-2-ene, 5-methylbicyclo[2.2.1]hept-2-ene, 5-ethylbicyclo[2.2.1]hept-2-ene, 5-butylbicyclo[2.2.1]hept-2-ene, 5-hexylbicyclo[2.2.1]hept-2-ene, 5-octylbicyclo[2.2.1]hept-2-ene, 5-hydroxybicyclo[2.2.1]hept-2-ene, (3-hydroxyadamantan-1-yl)bicyclo[2.2.1]hept-2-ene-5-carboxylate, (5(6)-hydroxybicyclo[2.2.1]hept-2-yl)bicyclo[2.2.1]hept-2-ene-5-carboxylate, (9(10)-hydroxytetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl)bicyclo[2.2.1]hept-2-ene-5-carboxylate, bicyclo[2.2.1]hept-2-ene-5-carboxylic acid, bicyclo[2.2.1]hept-2-ene-5-acetic acid, bicyclo[2.2.1]hept-2-ene-5-ethanecarboxylic acid, bicyclo[2.2.1]hept-2-ene-5-propionic acid, 5-cyanobicyclo[2.2.1]hept-2-ene, 5-cyanomethylbicyclo[2.2.1]hept-2-ene, 5-(2-cyanoethyl)bicyclo[2.2.1]hept-2-ene, 5-(3-cyanopropyl)bicyclo[2.2.1]hept-2-ene,

[0039]

t-butyl bicyclo[2.2.1]hept-2-ene-5-carboxylate, (2-methyl-2-propyl)bicyclo[2.2.1]hept-2-ene-5-carboxylate, (2-methyl-2-butyl, 2-ethyl-2-butyl)bicyclo[2.2.1]hept-2-ene-5-carboxylate, (3-ethyl-3-butyl)bicyclo[2.2.1]hept-2-ene-5-carboxylate, (2-methyladamantan-2-yl)bicyclo[2.2.1]hept-2-ene-5-carboxylate, (2-methyl-3-hydroxyadamantan-2-yl)bicyclo[2.2.1]hept-2-ene-5-carboxylate, (2-ethyladamantan-2-yl)bicyclo[2.2.1]hept-2-ene-5-carboxylate, (8-methyltricyclo[5.2.1.0^{2,6}]decan-8-yl)bicyclo[2.2.1]hept-2-ene-5-carboxylate, (8-ethyltricyclo[5.2.1.0^{2,6}]decan-8-yl)bicyclo[2.2.1]hept-2-ene-5-carboxylate, (1-methylcyclopentyl)bicyclo[2.2.1]hept-2-ene-5-carboxylate, (1-ethylcyclopentyl)bicyclo[2.2.1]hept-2-ene-5-carboxylate, (1-methylcyclohexyl)bicyclo[2.2.1]hept-2-ene-5-carboxylate, (1-ethylcyclohexyl)bicyclo[2.2.1]hept-2-ene-5-carboxylate, (2-methylbicyclo[2.2.1]hept-2-yl)

bicyclo[2.2.1]hept-2-ene-5-carboxylate, (2-ethylbicyclo[2.2.1]hept-2-yl)
 bicyclo[2.2.1]hept-2-ene-5-carboxylate, (4-methyltetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl)
 bicyclo[2.2.1]hept-2-ene-5-carboxylate, (4-ethyl-tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl)
 bicyclo[2.2.1]hept-2-ene-5-carboxylate,
 (1-cyclohexyl-1-methylethyl) bicyclo[2.2.1]hept-2-ene-5-carboxylate,
 (1-bicyclo[2.2.1]hept-2-yl-1-methylethyl) bicyclo[2.2.1]hept-2-ene-5-carboxylate,
 (1-tricyclo[5.2.1.0^{2,6}]dec-8-yl-1-methylethyl) bicyclo[2.2.1]hept-2-ene-5-carboxylate,
 (1-tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-yl-1-methylethyl)
 bicyclo[2.2.1]hept-2-ene-5-carboxylate, (1-adamantan-1-yl-1-methylethyl)
 bicyclo[2.2.1]hept-2-ene-5-carboxylate, (1-(2(3)-hydroxycyclopentyl)-1-methylethyl)
 bicyclo[2.2.1]hept-2-ene-5-carboxylate, (1-(3(4)-hydroxycyclohexyl)-1-methylethyl)
 bicyclo[2.2.1]hept-2-ene-5-carboxylate, (1-(3(4)-hydroxycycloheptyl)-1-methylethyl)
 bicyclo[2.2.1]hept-2-ene-5-carboxylate, (1-(3-hydroxyadamantan-1-yl)-1-methylethyl-1,1-dicyclohexylethyl) bicyclo[2.2.1]hept-2-ene-5-carboxylate,
 (1,1-dibicyclo[2.2.1]hept-2-ylethyl) bicyclo[2.2.1]hept-2-ene-5-carboxylate,
 (1,1-ditricyclo[5.2.1.0^{2,6}]dec-8-ylethyl) bicyclo[2.2.1]hept-2-ene-5-carboxylate,
 (1,1-di(tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl)ethyl) bicyclo[2.2.1]hept-2-ene-5-carboxylate,
 (1,1-diadamantan-1-ylethyl) bicyclo[2.2.1]hept-2-ene-5-carboxylate,

[0040]

methyl bicyclo[2.2.1]hept-2-ene-5-carboxylate,
 ethyl bicyclo[2.2.1]hept-2-ene-5-carboxylate, n-propyl
 bicyclo[2.2.1]hept-2-ene-5-carboxylate, cyclopentyl bicyclo[2.2.1]hept-2-ene-5-carboxylate,
 cyclohexyl bicyclo[2.2.1]hept-2-ene-5-carboxylate, (adamantan-1-yl)
 bicyclo[2.2.1]hept-2-ene-5-carboxylate, (bicyclo[2.2.1]hept-2-yl)
 bicyclo[2.2.1]hept-2-ene-5-carboxylate, (7,7-dimethylbicyclo[2.2.1]hept-1-yl)
 bicyclo[2.2.1]hept-2-ene-5-carboxylate, (tricyclo[5.2.1.0^{2,6}]dec-8-yl)
 bicyclo[2.2.1]hept-2-ene-5-carboxylate, (tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-yl)
 bicyclo[2.2.1]hept-2-ene-5-carboxylate;

[0041]

(5-oxo-4-oxatricyclo[4.2.1.0^{3,7}]nonan-2-yl) bicyclo[2.2.1]hept-2-ene-5-carboxylate,
 (9-methoxycarbonyl-5-oxo-4-oxatricyclo[4.2.1.0^{3,7}]nonan-2-yl)
 bicyclo[2.2.1]hept-2-ene-5-carboxylate, (7-oxo-6-oxabicyclo[3.2.1]octan-4-yl)
 bicyclo[2.2.1]hept-2-ene-5-carboxylate,
 (2-methoxycarbonyl-7-oxo-6-oxabicyclo[3.2.1]octan-4-yl)
 bicyclo[2.2.1]hept-2-ene-5-carboxylate, (2-oxotetrahydropyran-4-yl)
 bicyclo[2.2.1]hept-2-ene-5-carboxylate, (4-methyl-2-oxotetrahydropyran-4-yl)
 bicyclo[2.2.1]hept-2-ene-5-carboxylate, (4-ethyl-2-oxotetrahydropyran-4-yl)
 bicyclo[2.2.1]hept-2-ene-5-carboxylate, (4-propyl-2-oxotetrahydropyran-4-yl)
 bicyclo[2.2.1]hept-2-ene-5-carboxylate, (5-oxotetrahydrofuran-3-yl)
 bicyclo[2.2.1]hept-2-ene-5-carboxylate, (2,2-dimethyl-5-oxotetrahydrofuran-3-yl)
 bicyclo[2.2.1]hept-2-ene-5-carboxylate, (4,4-dimethyl-5-oxotetrahydrofuran-3-yl)

bicyclo[2.2.1]hept-2-ene-5-carboxylate, (2-oxotetrahydrofuran-3-yl)
 bicyclo[2.2.1]hept-2-ene-5-carboxylate, (4,4-dimethyl-2-oxotetrahydrofuran-3-yl)
 bicyclo[2.2.1]hept-2-ene-5-carboxylate, (5,5-dimethyl-2-oxotetrahydrofuran-3-yl)
 bicyclo[2.2.1]hept-2-ene-5-carboxylate, (2-oxotetrahydrofuran-3-yl)
 bicyclo[2.2.1]hept-2-ene-5-carboxylate, (5-oxotetrahydrofuran-2-ylmethyl)
 bicyclo[2.2.1]hept-2-ene-5-carboxylate, (3,3-dimethyl-5-oxotetrahydrofuran-2-ylmethyl)
 bicyclo[2.2.1]hept-2-ene-5-carboxylate, (4,4-dimethyl-5-oxotetrahydrofuran-2-ylmethyl)
 bicyclo[2.2.1]hept-2-ene-5-carboxylate,
 9-hydroxymethyltetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene,
 9-(2-hydroxyethyl)tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene,
 9-(3-hydroxypropyl)tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene,
 9-(1-fluoro-1-hydroxymethyl)tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene,
 9-(1,1-fluoro-1-hydroxymethyl)tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene,
 9-(1,2-difluoro-2-hydroxymethyl)tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene,
 9-(1,1,2,2-tetrafluoro-2-hydroxymethyl)tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene,
 9-(2-trifluoromethyl-2-hydroxyethyl)tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene,
 9-(2,2-ditrifluoromethyl-2-hydroxyethyl)tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene,

[0042]

tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene, 9-methyltetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene,
 9-ethyltetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene, 9-butyltetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene,
 9-hexyltetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene, 9-octyltetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene,
 9-hydroxytetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene, (3-hydroxyadamantan-1-yl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate, (5(6)-hydroxybicyclo[2.2.1]hept-2-yl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate,
 (9(10)-hydroxytetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-yl]
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate,
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylic acid,
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-acetic acid,
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-ethane carboxylic acid,
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-propionic acid,
 9-cyanotetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene,
 9-cyanomethyltetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene,
 9-(2-cyanoethyl)tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene,
 9-(3-cyanopropyl)tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene,

[0043]

t-butyl tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate, (2-methyl-2-propyl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate, (2-methyl-2-butyl, 2-ethyl-2-butyl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate, (3-ethyl-3-butyl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate, (2-methyladamantan-2-yl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate, (2-methyl-3-hydroxyadamantan-2-yl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate, (2-ethyladamantan-2-yl)

tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate, (8-methyltricyclo[5.2.1.0^{2,6}]decan-8-yl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate, (8-ethyltricyclo[5.2.1.0^{2,6}]decan-8-yl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate, (1-methylcyclopentyl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate, (1-ethylcyclopentyl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate, (1-methylcyclohexyl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate, (1-ethylcyclohexyl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate, (2-methylbicyclo[2.2.1]hept-2-yl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate, (2-ethylbicyclo[2.2.1]hept-2-yl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate,
 (4-methyltetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-yl]
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate,
 (4-ethyltetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-yl]
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate, (1-cyclohexyl-1-methylethyl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate,
 (1-bicyclo[2.2.1]hept-2-yl-1-methylethyl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate,
 (1-tricyclo[5.2.1.0^{2,6}]dec-8-yl-1-methylethyl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate,
 (1-tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-yl-1-methylethyl]
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate, (1-adamantan-1-yl-1-methylethyl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate,
 (1-(2(3)-hydroxycyclopentyl)-1-methylethyl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate,
 (1-(3(4)-hydroxycyclohexyl)-1-methylethyl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate,
 (1-(3(4)-hydroxycycloheptyl)-1-methylethyl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate,
 (1-(3-hydroxyadamantan-1-yl)-1-methylethyl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate, (1,1-dicyclohexylethyl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate, (1,1-dibicyclo[2.2.1]hept-2-ylethyl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate, (1,1-ditricyclo[5.2.1.0^{2,6}]dec-8-ylethyl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate,
 (1,1-di(tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-yl)ethyl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate, (1,1-diadamantan-1-ylethyl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate,

[0044]

methyl tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate, ethyl
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate, n-propyl
 bicyclo[2.2.1]hept-2-ene-5-carboxylate, cyclopentyl
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate, cyclohexyl
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate, (adamantan-1-yl)

tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate, (bicyclo[2.2.1]hept-2-yl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate, (7,7-dimethylbicyclo[2.2.1]hept-1-yl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate, (tricyclo[5.2.1.0^{2,6}]dec-8-yl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate, (tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-yl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate;

[0045]

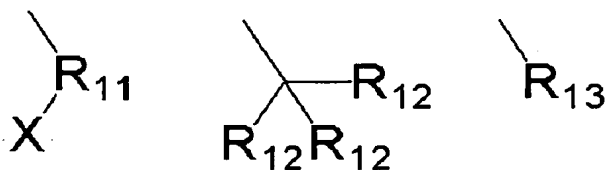
(5-oxo-4-oxa-tricyclo[4.2.1.0^{3,7}]non-2-yl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate,
 (9-methoxycarbonyl-5-oxo-4-oxa-tricyclo[4.2.1.0^{3,7}]non-2-yl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate, (7-oxo-6-oxabicyclo[3.2.1]oct-4-yl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate,
 (2-methoxycarbonyl-7-oxo-6-oxabicyclo[3.2.1]oct-4-yl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate, (2-oxotetrahydropyran-4-yl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate, (4-methyl-2-oxotetrahydropyran-4-yl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate, (4-ethyl-2-oxotetrahydropyran-4-yl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate, (4-propyl-2-oxotetrahydropyran-4-yl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate, (5-oxotetrahydrofuran-3-yl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate,
 (2,2-dimethyl-5-oxotetrahydrofuran-3-yl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate,
 (4,4-dimethyl-5-oxotetrahydrofuran-3-yl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate, (2-oxotetrahydrofuran-3-yl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate,
 (4,4-dimethyl-2-oxotetrahydrofuran-3-yl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate,
 (5,5-dimethyl-2-oxotetrahydrofuran-3-yl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate, (2-oxotetrahydrofuran-3-yl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate, (5-oxotetrahydrofuran-2-ylmethyl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate,
 (3,3-dimethyl-5-oxotetrahydrofuran-2-ylmethyl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate,
 (4,4-dimethyl-5-oxotetrahydrofuran-2-ylmethyl)
 tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-ene-9-carboxylate, and the like.

[0046]

The substituent R₃ constituting the repeating unit represented by the above formula (IV) (hereinafter referred to as "repeating unit (3)") include ones as mentioned above as examples of the substituent R₁ in the repeating unit (1).

[0047]

Additionally, examples of the substituent R₄ constituting the above repeating unit (3) include organic groups as follows.



(4-1)

(4-2)

(4-3)

(In the organic group (4-1), R_{11} may represent a divalent organic group having a linear, branched, or cyclic skeleton, in which case, X represents hydrogen atom, hydroxyl group, carboxyl group, nitro group, cyano group, or amino group, or R_{11} may not be present, in which case, X represents a carboxyl group or a cyano group. In the organic group (4-2), R_{12} individually represents an alicyclic hydrocarbon group having 4-20 carbon atoms or a derivative thereof, or an alkyl group having 1-4 carbon atoms. In the organic group (4-3), R_{13} represents an alkyl group having 1-6 carbon atoms or a derivative thereof, a cycloalkyl group having 5-10 carbon atoms or a derivative thereof, a polyalicyclic hydrocarbon group having 4-20 carbon atoms or a derivative thereof, or a lactone ring.)

[0048]

The substituent R_{11} in the organic group (4-1) includes groups mentioned above as examples of R_5 in the above organic group (2-2). The substituent R_{12} in the organic group (4-2) includes groups mentioned above as examples of R_6 in the above organic group (2-3). In addition, the substituent R_{13} in the organic group (4-3) includes groups mentioned above as examples of the substituent R_7 in the above organic group (2-4).

[0049]

Examples of monomer providing the repeating unit (3) are as follows:
 hydroxymethyl (meth)acrylate, 2-hydroxymethyl 1-(meth)acrylate, 3-hydroxypropyl 1-(meth)acrylate, 1-fluoro-1-hydroxymethyl 1-(meth)acrylate, 1,1-difluoro-1-hydroxymethyl 1-(meth)acrylate, 1, 2-difluoro-2-hydroxymethyl 1-(meth)acrylate, 1,1,2,2-tetrafluoro-2-hydroxymethyl 1-(meth)acrylate, 2-trifluoromethyl-2-hydroxyethyl 1-(meth)acrylate, 2,2-ditrifluoromethyl-2-hydroxyethyl 1-(meth)acrylate,

[0050]

(meth)acrylic acid, 3-hydroxyadamant-1-yl (meth)acrylate, 5(6)-hydroxybicyclo[2.2.1]hept-2-yl (meth)acrylate, 9(10)-hydroxytetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-yl (meth)acrylate, carboxymethyl (meth)acrylate, 2-carboxyethyl (meth)acrylate, 3-carboxypropyl (meth)acrylate, 3-carboxyadamant-1-yl (meth)acrylate, 5(6)-carboxybicyclo[2.2.1]hept-2-yl (meth)acrylate, 9(10)-carboxytetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-yl (meth)acrylate, cyanomethyl (meth)acrylate, 2-cyanoethyl 1-(meth)acrylate, 3-cyanopropyl 1-(meth)acrylate, 3-cyanoadamantan-1-yl (meth)acrylate, 5(6)-cyanobicyclo[2.2.1]hept-2-ylethyl (meth)acrylate, 9(10)-cyanotetracyclotetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-yl (meth)acrylate,

[0051]

t-butyl (meth)acrylate, 2-methyl-2-propyl (meth)acrylate, 2-methyl-2-butyl

(meth)acrylate, 2-ethyl-2-butyl (meth)acrylate, 3-ethyl-3-butyl (meth)acrylate, 2-methyladamantan-2-yl (meth)acrylate, 2-methyl-3-hydroxyadamantan-2-yl (meth)acrylate, 2-ethyladamantan-2-yl (meth)acrylate, 8-methyltricyclo[5.2.1.0^{2,6}]decan-8-yl (meth)acrylate, 8-ethyltricyclo[5.2.1.0^{2,6}]decan-8-yl (meth)acrylate, 1-methylcyclopentyl (meth)acrylate, 1-ethylcyclopentyl (meth)acrylate, 1-methylcyclohexyl (meth)acrylate, 1-ethylcyclohexyl (meth)acrylate, 2-methylbicyclo[2.2.1]hept-2-yl (meth)acrylate, 2-ethylbicyclo[2.2.1]hept-2-yl (meth)acrylate, 4-methyltetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-yl (meth)acrylate, 4-ethyltetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-yl (meth)acrylate, 1-cyclohexyl-1-methylethyl (meth)acrylate, 1-bicyclo[2.2.1]hept-2-yl-1-methylethyl(meth)acrylate, 1-tricyclo[5.2.1.0^{2,6}]dec-8-yl-1-methylethyl (meth)acrylate, 1-tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-yl-1-methylethyl (meth)acrylate, 1-adamantan-1-yl-1-methylethyl (meth)acrylate, 1-(2(3)-hydroxycyclopentyl)-1-methylethyl (meth)acrylate, 1-(3(4)-hydroxycyclohexyl)-1-methylethyl (meth)acrylate, 1-(3(4)-hydroxycycloheptyl)-1-methylethyl (meth)acrylate, 1-(3-hydroxyadamantan-1-yl)-1-methylethyl (meth)acrylate, 1,1-dicyclohexylethyl (meth)acrylate, 1,1-dibicyclo[2.2.1]hept-2-ylethyl (meth)acrylate, 1,1-ditricyclo[5.2.1.0^{2,6}]dec-8-ylethyl (meth)acrylate, 1,1-di(tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-yl)ethyl (meth)acrylate, 1,1-diadamantan-1-yl-1-ethyl (meth)acrylate,

[0052]

methyl (meth)acrylate, ethyl (meth)acrylate, n-propyl (meth)acrylate, cyclopentyl (meth)acrylate, cyclohexyl (meth)acrylate, adamantan-1-yl (meth)acrylate, bicyclo[2.2.1]hept-2-yl (meth)acrylate, 7,7-dimethylbicyclo[2.2.1]hept-1-yl (meth)acrylate, tricyclo[5.2.1.0^{2,6}]dec-8-yl (meth)acrylate, tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodec-4-yl (meth)acrylate,

[0053]

5-oxo-4-oxatricyclo[4.2.1.0^{3,7}]non-2-yl (meth)acrylate, 9-methoxycarbonyl-5-oxo-4-oxa-tricyclo[4.2.1.0^{3,7}]non-2-yl (meth)acrylate, 7-oxo-6-oxabicyclo[3.2.1]octan-4-yl (meth)acrylate, 2-methoxycarbonyl-7-oxo-6-oxa-bicyclo[3.2.1]oct-4-yl (meth)acrylate, 2-oxotetrahydropyran-4-yl (meth)acrylate, 4-methyl-2-oxotetrahydropyran-4-yl (meth)acrylate, 4-ethyl-2-oxotetrahydropyran-4-yl (meth)acrylate, 4-propyl-2-oxotetrahydropyran-4-yl (meth)acrylate, 5-oxotetrahydrofuran-3-yl (meth)acrylate, 2,2-dimethyl-5-oxotetrahydrofuran-3-yl (meth)acrylate, 4,4-dimethyl-5-oxotetrahydrofuran-3-yl (meth)acrylate, 2-oxotetrahydrofuran-3-yl (meth)acrylate, 4,4-dimethyl-2-oxotetrahydrofuran-3-yl (meth)acrylate, 5,5-dimethyl-2-oxotetrahydrofuran-3-yl (meth)acrylate, 2-oxotetrahydrofuran-3-yl (meth)acrylate, 5-oxotetrahydrofuran-2-yl (meth)acrylate, 3,3-dimethyl-5-oxotetrahydrofuran-2-yl (meth)acrylate, 4,4-dimethyl-5-oxotetrahydrofuran-2-yl (meth)acrylate, and the like.

[0054]

Either one type or two or more types of the above repeating units (2) and (3) may be contained in the resin [A].

[0055]

The total amount of the repeating units (2) and/or (3) is usually 80 mol% or less, preferably 70 mol% or less, and still more preferably 60 mol% or less of the total amount of the repeating units. If the content of the repeating units (2) and/or (3) is more than 80 mol%, not only does adhesion to the substrate and developability tend to decrease, but also the effect of reducing development defects tends to be impaired.

[0056]

Further, a monomer unit constituting the resin [A] may further be one formed by a monomer as follows, and the like. For example, a monofunctional monomer including a vinyl ester such as vinyl acetate, vinyl propionate and vinyl butyrate; an unsaturated nitrile compound such as (meth)acrylonitrile, α -chloroacrylonitrile, crotonitrile, maleinitrile, fumaronitrile, mesaconitrile, citraconitrile, and itaconitrile; an unsaturated amide compound such as (meth)acrylamide, N,N-dimethyl(meth)acrylamide, crotonamide, maleinamide, fumaramide, mesaconamide, citraconamide and itaconamide; other nitrogen-containing vinyl compounds such as N-vinyl- ϵ -caprolactam, N-vinylpyrrolidone, vinylpyridine and vinylimidazole; an unsaturated carboxylic acid (anhydride) such as crotonic acid, maleic acid, maleic anhydride, fumaric acid, itaconic acid, itaconic anhydride, citraconic acid, citraconic anhydride and mesaconic acid; and the like;

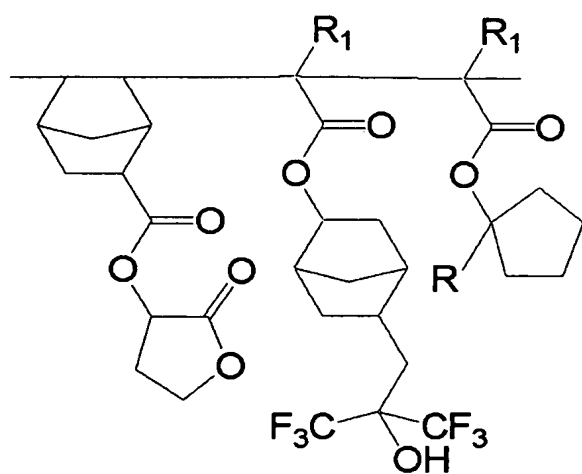
a polyfunctional monomer such as methylene glycol di(meth)acrylate, ethylene glycol di(meth)acrylate, propylene glycol di(meth)acrylate, 1,6-hexanediol di(meth)acrylate, 2,5-dimethyl-2,5-hexanediol di(meth)acrylate, 1,8-octanediol di(meth)acrylate, 1,9-nonanediol di(meth)acrylate, 1,4-bis(2-hydroxypropyl)benzene di(meth)acrylate, 1,3-bis(2-hydroxypropyl)benzene di(meth)acrylate, 1,2-adamantanediol di(meth)acrylate, 1,3-adamantanediol di(meth)acrylate, 1,4-adamantanediol di(meth)acrylate and tricyclodecanyldimethylol di(meth)acrylate; may be used.

Either one type or two or more types of these repeating units may be contained in the resin [A].

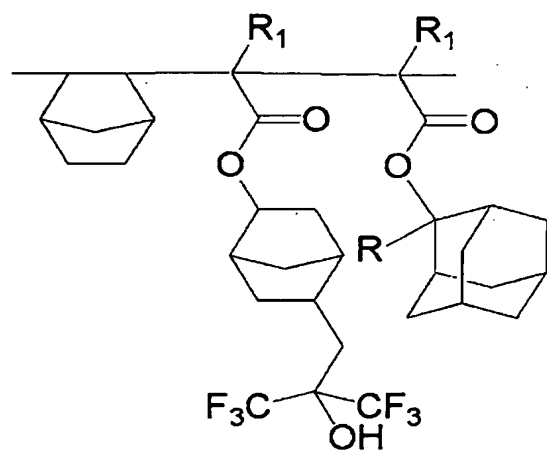
Additionally, the content of these repeating units is usually 20 mol% or less, and preferably 10 mol% or less based on the total content of the repeating units.

[0057]

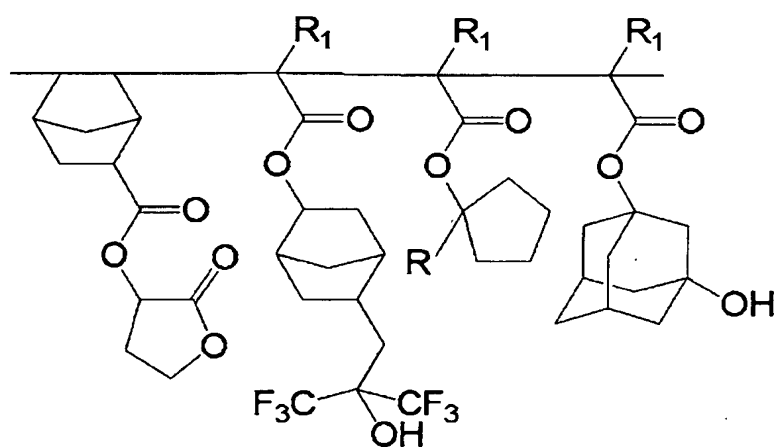
Examples of the resin [A] according to the present invention include a resin having the following monomer unit described below. There are no specific limitations to the order and proportion of the monomer units constituting the resin [A].



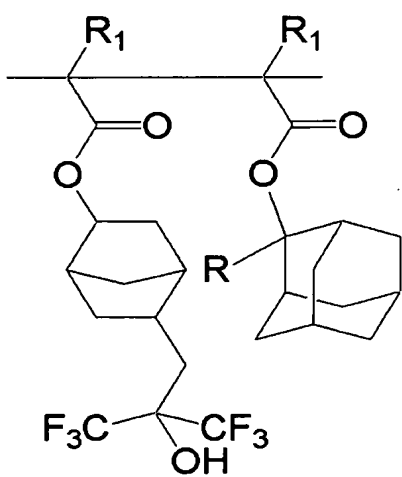
[0058]



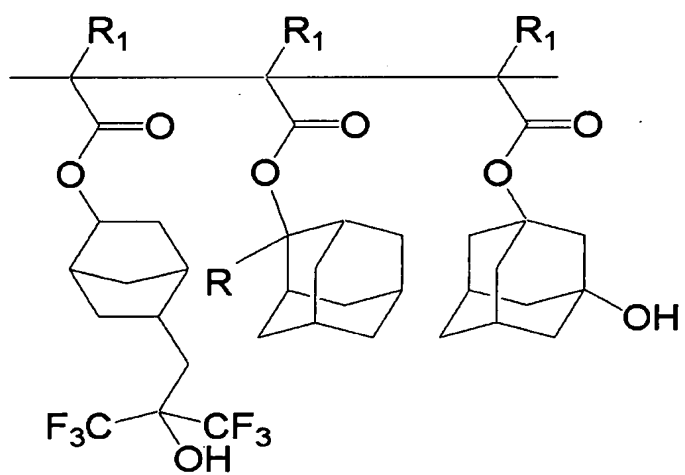
[0059]



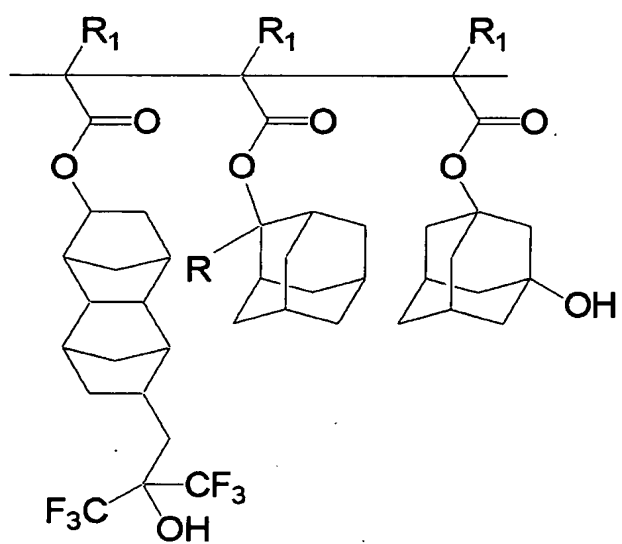
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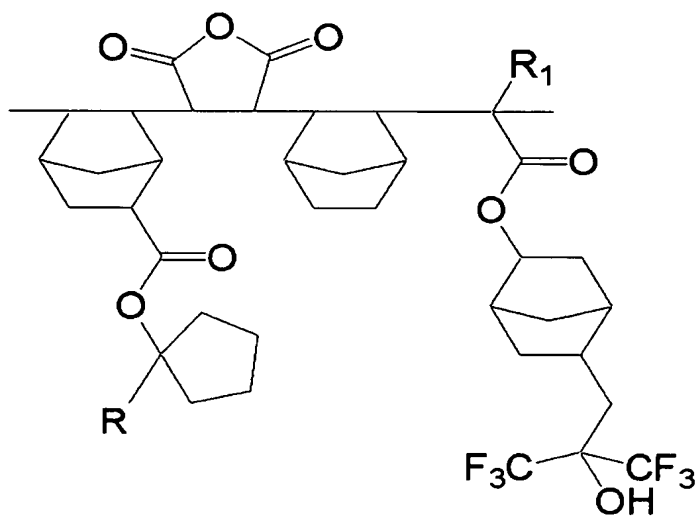
[0061]



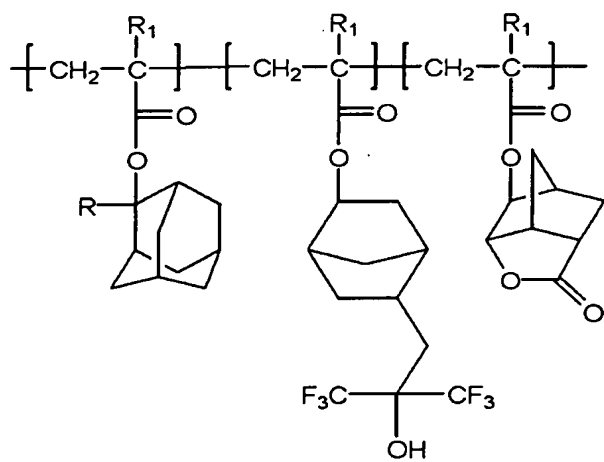
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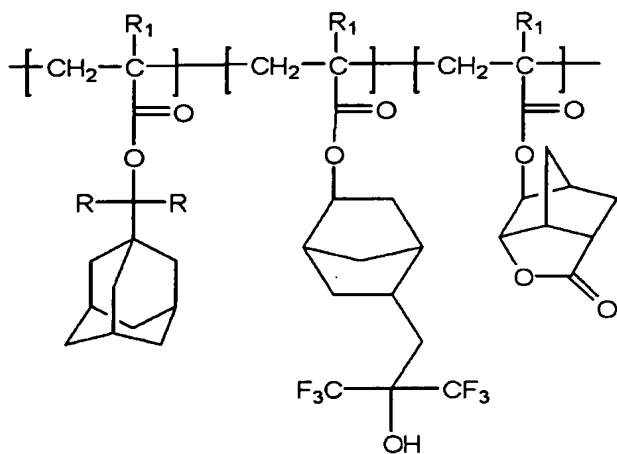
[0063]



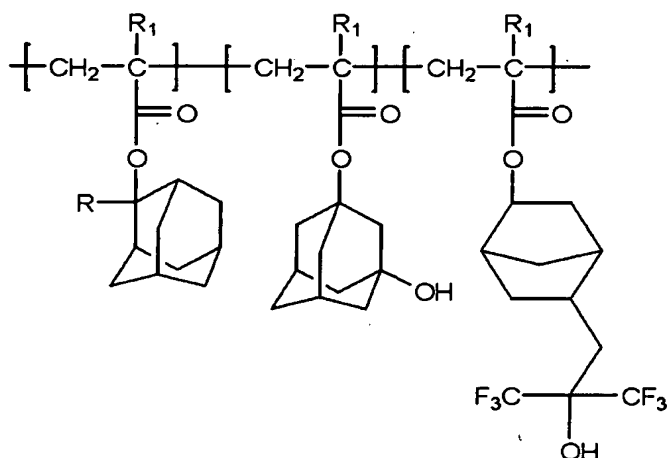
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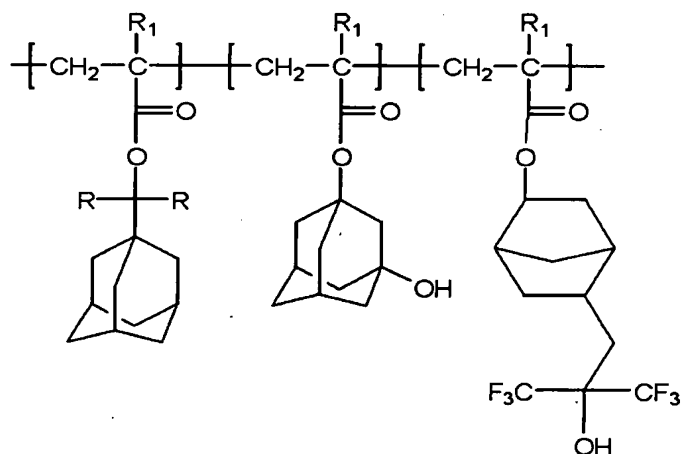
[0065]



[0066]



[0067]



(In all the above formulas, R₁ represents hydrogen atom, methyl group, a hydroxylalkyl group having 1-4 carbon atoms, or a perfluoroalkyl group having 1-4 carbon atoms and R represents methyl group or ethyl group.)

[0068]

The polystyrene-reduced weight-average molecular weight (hereinafter referred to as "Mw") of the resin [A] determined by gel permeation chromatography (GPC) is usually 1,000-300,000, preferably 2,000-200,000, and more preferably 3,000-100,000. If the Mw is less than 1,000, heat resistance as a resist may decrease. If the Mw exceeds 300,000, developability as a resist may decrease.

In addition, the ratio of Mw to the number average molecular weight (hereinafter referred to as "Mn") determined at the same time as the Mw (Mw/Mn) by GPC is usually 1-5, and preferably 1-3.

The resin [A] contained in the radiation-sensitive resin composition of the present invention may be used alone or in combinations of two or more.

[0069]

The resin [A] is prepared by polymerizing a mixture of monomers providing each of the above repeating units in an appropriate solvent in the presence of a chain transfer agent, as required, using a radical polymerization initiator such as a hydroperoxide, a dialkyl peroxide, a diacyl peroxide and an azo compound.

Examples of the solvent which is be used for the polymerization include an alkane such as n-pentane, n-hexane, n-heptane, n-octane, n-nonane and n-decane; a cycloalkane such as cyclohexane, cycloheptane, cyclooctane, decalin and norbornane; an aromatic hydrocarbon such as benzene, toluene, xylene, ethylbenzene and cumene; a halogenated hydrocarbon such as chlorobutanes, bromohexanes, dichloroethanes, hexamethylene dibromide and chlorobenzene; a saturated carboxylic acid ester such as ethyl acetate, n-butyl acetate, i-butyl acetate, methyl propionate and propylene glycol monomethyl ether acetate; an alkyl lactone such as γ -butyrolactone; an ether such as tetrahydrofuran, dimethoxyethanes and diethoxyethanes; an alkyl ketone such as 2-butanone, 2-heptanone, and methyl isobutyl ketone; a cycloalkyl ketone such as cyclohexanone; an alcohol such as 2-propanol and propylene glycol monomethyl ether; and the like. These solvents may be used alone or in combinations of two or more.

[0070]

Further, the polymerization temperature is usually in the range from 40 to 120°C and preferably from 50 to 100°C. The reaction time is usually in the range from 1 to 48 hours and preferably from 1 to 24 hours.

[0071]

It is preferable that the resin [A] according to the present invention has a high purity and that not only the resin is almost free from impurities such as halogens or metals, but also the content of residual monomers and oligomers should be less than the prescribed amount, for example, the content determined by HPLC should be 0.1 wt% or less. Satisfying these requirements not only improves sensitivity, resolution, process stability, and pattern configuration of the resist obtained from the radiation-sensitive resin composition of the present invention even more, but ensures a resist with only a small change in foreign matter content and sensitivity over time.

As examples of a purification method for the resin [A] thus obtained, the following methods can be given. As a method for removing impurities such as metals, a method of putting metals in the polymer solution to be adsorbed using a zeta-potential filter, a method of causing the metals to be in a chelate state by washing the polymer solution with an acidic aqueous solution such as oxalic acid or sulfonic acid and removing the metals, and the like can be given. As a method for removing the residual monomers and oligomer components to a content less than a specific level, a liquid-liquid extraction method in which the residual monomers and oligomer components are removed by combining washing with water and a suitable solvent, a purification method in a liquid state such as ultrafiltration in which only the monomers and oligomer components with a molecular weight less than a specific value are extracted and removed, a reprecipitation method in which the residual monomers and the like

are removed by adding the polymer solution to a poor solvent dropwise, thereby causing the polymer solution to coagulate in the poor solvent, and a purification method in a solid state in which the resin slurry separated by filtration is washed with a poor solvent can be given. These methods may be used in combination.

[0072]

2. Radiation-sensitive acid generator [B]

The radiation-sensitive acid generator [B] contained in the radiation-sensitive resin composition of the present invention (hereinafter referred to as "acid generator [B]") is a compound which generates an acid upon exposure to radiation such as visible rays, ultraviolet rays, deep ultraviolet rays, electron beams and X-rays.

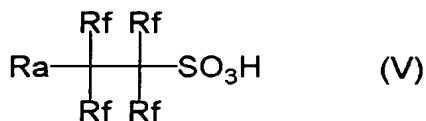
The acid generator [B] causes an acid-dissociating group such as an alkyl adamantyl group, t-butyl group, or tetrahydropyranyl group in the resin [A] to dissociate by the action of the acid generated upon exposure to radiation. As a result, the exposed area on the resist film is rendered soluble in an alkaline developer to form a positive-tone resist pattern. The acid generator [B] consists of a parent nucleus and a generated acid.

[0073]

2-1. Generated acid

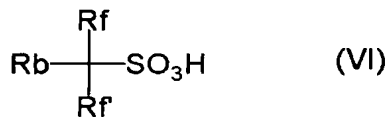
Examples of the generated acid include a sulfonic acid compound, a carboxylic-acid compound, an imide compound and the like. Specific compounds include the compounds represented by the following formulas (V) to (IX).

[0074]



(In the formula, Ra represents hydrogen atom, fluorine atom, an alkyl group having 1-20 carbon atoms, a fluoroalkyl group having 1-20 carbon atoms, a cyclic hydrocarbon group having 3-20 carbon atoms or a cyclic fluoro-hydrocarbon group having 3-20 carbon atoms, and the cyclic hydrocarbon group and the cyclic fluoro-hydrocarbon group may have a substituent. Rf represents fluorine atom or trifluoromethyl group.)

[0075]



(In the formula, Rb represents hydrogen atom, an alkyl group having 1-20 carbon atoms, a cyclic hydrocarbon group having 3-20 carbon atoms or a cyclic fluoro-hydrocarbon group having 3-20 carbon atoms, and the cyclic hydrocarbon group and the cyclic fluoro-hydrocarbon group may have a substituent. Rf represents fluorine atom or

trifluoromethyl group, and Rf is hydrogen atom, fluorine atom, methyl group, or trifluoromethyl group.)

[0076]



(In the formula, Rs represents an alkyl group having 1-20 carbon atoms or a cyclic hydrocarbon group having 3-20 carbon atoms, and the cyclic hydrocarbon group may have a substituent.)

[0077]



(In the formula, Rc represents an alkyl group having 1-20 carbon atoms, a fluoroalkyl group having 1-20 carbon atoms, a cyclic hydrocarbon group having 3-20 carbon atoms or a cyclic fluoro-hydrocarbon group having 3-20 carbon atoms, and the cyclic hydrocarbon group and the cyclic fluoro-hydrocarbon group may have a substituent.)

[0078]



(In the formula, Re represents a Rg-SO₂- group or a Rg-CO- group, and Rg is the same group as Ra mentioned above.)

[0079]

When each of the substituent Ra in the formula (V), the substituent Rb in the formula (VI), the substituent Rs in the formula (VII), the substituent Rc in the formula (VIII) and the substituent Rg constituting the substituent Re in the formula (IX) is an alkyl group, the alkyl group may be either linear or branched. Examples of the alkyl group include methyl group, ethyl group, n-propyl group, i-propyl group, n-butyl group, i-butyl group, sec-butyl group, t-butyl group, n-pentyl group, n-hexyl group, n-heptyl group, n-octyl group, 2-ethylhexyl group, nonyl group, decyl group, dodecyl group and the like.

When each of the substituent Ra in the formula (V), the substituent Rc in the formula (VIII) and the substituent Rg constituting the substituent Re in the formula (IX) is a fluoroalkyl group, the fluoroalkyl group may be either linear or branched. Examples of the fluoroalkyl group include trifluoromethyl group, pentafluoroethyl group,

heptafluoro-n-propyl group, heptafluoro-i-propyl group, nonafluoro-n-butyl group, nonafluoro-i-butyl group, nonafluoro-sec-butyl group, nonafluoro-t-butyl group, perfluoro-n-pentyl group, perfluoro-n-hexyl group, perfluoro-n-heptyl group, perfluoro-n-octyl group and the like.

[0080]

When each of the substituent Ra in the formula (V), the substituent Rb in the formula (VI), the substituent Rs in the formula (VII), the substituent Rc in the formula (VIII) and the substituent Rg constituting the substituent Re in the formula (IX) is a cyclic hydrocarbon group, the cyclic hydrocarbon group may be either an alicyclic hydrocarbon group or a hydrocarbon group containing an aromatic ring. Examples of the cyclic hydrocarbon group include cyclopropyl group, cyclobutyl group, cyclopentyl group, cycloheptyl group, cyclooctyl group, phenyl group, tolyl group, xylyl group, benzyl group, naphthyl group and the like. In addition, when each of the substituent Ra in the formula (V), the substituent Rb in the formula (VI), the substituent Rc in the formula (VIII) and the substituent Rg constituting the substituent Re in the formula (IX) is a cyclic fluoro-hydrocarbon group, the cyclic fluoro-hydrocarbon group may be either an alicyclic hydrocarbon group or a hydrocarbon group comprising an aromatic ring. Examples of the cyclic fluoro-hydrocarbon group include a group in which at least one hydrogen atom of the above-mentioned cyclic hydrocarbon group is replaced with fluorine atom.

[0081]

When the cyclic hydrocarbon group and cyclic fluoro-hydrocarbon group mentioned above have further a substituent, examples of the substituent group include phenyl group, 2-naphthyl group, a cycloalkyl group, bicyclo[2.2.1]heptan-2-yl group, tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl group, adamantan-1-yl group, camphor group and derivatives thereof, and the like.

[0082]

Accordingly, specific examples of the acid represented by the above formula (V) include a linear or branched fluoroalkyl sulfonic acid, a 2-alicyclic skeleton substitute having an alicyclic skeleton at the 2-position and the like.

Examples of the linear or branched fluoroalkyl sulfonic acid include trifluoromethanesulfonic acid, pentafluoroethanesulfonic acid, heptafluoro-n-propanesulfonic acid, nonafluoro-n-butanesulfonic acid, perfluoro-n-octanesulfonic acid, 1,1,2,2-tetrafluoro-n-propanesulfonic acid, 1,1,2,2-tetrafluoro-n-butanesulfonic acid, 1,1,2,2-tetrafluoro-n-octanesulfonic acid and the like.

[0083]

Examples of the 2-alicyclic skeleton substitute having an alicyclic skeleton at the 2-position include a 1,1,2,2-tetrafluoroethanesulfonic acid, a 1-trifluoromethyl-1,2,2-trifluoroethanesulfonic acid, a 2-trifluoromethyl-1,1,2-trifluoroethanesulfonic acid, a 1,2-ditrifluoromethyl-1,2-difluoroethanesulfonic acid, a 1,1-ditrifluoromethyl-2,2-difluoroethanesulfonic acid, a

2,2-ditrifluoromethyl-1,1-difluoroethanesulfonic acid and the like.

[0084]

Further, examples of the 1,1,2,2-tetrafluoroethanesulfonic acid having an alicyclic skeleton at the 2-position include 2-cyclobutyl-1,1,2,2-tetrafluoroethanesulfonic acid, 2-cyclopentyl-1,1,2,2-tetrafluoroethanesulfonic acid, 2-cyclohexyl-1,1,2,2-tetrafluoroethanesulfonic acid, 2-phenyl-1,1,2,2-tetrafluoroethanesulfonic acid, 2-(4-trifluoromethylphenyl)-1,1,2,2-tetrafluoroethanesulfonic acid, 2-(2,3-difluorophenyl)-1,1,2,2-tetrafluoroethanesulfonic acid, 2-(2,4-difluorophenyl)-1,1,2,2-tetrafluoroethanesulfonic acid, 2-(2,5-difluorophenyl)-1,1,2,2-tetrafluoroethanesulfonic acid, 2-(2,6-difluorophenyl)-1,1,2,2-tetrafluoroethanesulfonic acid, 2-(3,4-difluorophenyl)-1,1,2,2-tetrafluoroethanesulfonic acid, 2-(3,5-difluorophenyl)-1,1,2,2-tetrafluoroethanesulfonic acid, 2-(3,6-difluorophenyl)-1,1,2,2-tetrafluoroethanesulfonic acid, 2-(2,3,4,5,6-pentafluorophenyl)-1,1,2,2-tetrafluoroethanesulfonic acid, 2-naphthalen-1-yl-1,1,2,2-tetrafluoroethanesulfonic acid, 2-naphthalen-2-yl-1,1,2,2-tetrafluoroethanesulfonic acid, 2-bicyclo[2.2.1]heptan-2-yl-1,1,2,2-tetrafluoroethanesulfonic acid, 2-(5-hydroxybicyclo[2.2.1]heptan-2-yl)-1,1,2,2-tetrafluoroethanesulfonic acid, 2-(6-hydroxybicyclo[2.2.1]heptan-2-yl)-1,1,2,2-tetrafluoroethanesulfonic acid, 2-(7,7-dimethylbicyclo[2.2.1]heptan-2-yl)-1,1,2,2-tetrafluoroethanesulfonic acid, 2-(tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl)-1,1,2,2-tetrafluoroethanesulfonic acid, 2-(9-hydroxytetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl)-1,1,2,2-tetrafluoroethanesulfonic acid, 2-(10-hydroxytetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl)-1,1,2,2-tetrafluoroethanesulfonic acid, 2-adamantan-1-yl-1,1,2,2-tetrafluoroethanesulfonic acid, 2-(1-adamantyl)-1,1,2,2-tetrafluoroethanesulfonic acid, 2-adamantan-2-yl-1,1,2,2-tetrafluoroethanesulfonic acid, 2-(3-hydroxyadamantan-1-yl)-1,1,2,2-tetrafluoroethanesulfonic acid, 2-(3-hydroxyadamantan-2-yl)-1,1,2,2-tetrafluoroethanesulfonic acid and the like.

[0085]

Specific examples of the acid represented by the above formula (VI) include a linear or branched fluoroalkyl sulfonic acid, an alicyclic skeleton substitute having an alicyclic skeleton at the 1- or 2-position and the like.

Examples of the linear or branched fluoroalkyl sulfonic acid include 1-fluoroethanesulfonic acid, 1-fluoro-n-propanesulfonic acid, 1-fluoro-n-butanesulfonic acid, 1-fluoro-n-octanesulfonic acid, 1,1-difluoroethanesulfonic acid, 1,1-difluoro-n-propanesulfonic acid, 1,1-difluoro-n-butanesulfonic acid, 1,1-difluoro-n-octanesulfonic acid, 1-trifluoromethyl-n-propanesulfonic acid, 1-trifluoromethyl-n-butanesulfonic acid, 1-trifluoromethyl-n-octanesulfonic acid, 1,1-bis(trifluoromethyl)ethanesulfonic acid, 1,1-bis(trifluoromethyl)-n-propanesulfonic acid,

1,1-bis(trifluoromethyl)-n-butanesulfonic acid, 1,1-bis(trifluoromethyl)-n-octanesulfonic acid and the like.

[0086]

Examples of the alicyclic skeleton substitute having an alicyclic skeleton at the 1- or 2-position include an alicyclic skeleton substitute of a 1,1-difluoromethanesulfonic acid, a 1,1-difluoroethanesulfonic acid, a 1-monofluoromethanesulfonic acid, a 1-monofluoroethanesulfonic acid, a 1-trifluoromethylmethanesulfonic acid, a 1-trifluoromethylethanesulfonic acid, a 1,1-ditrifluoromethylmethanesulfonic acid, a 1,1-ditrifluoromethylethanesulfonic acid and the like.

[0087]

Examples of the 1,1-difluoromethanesulfonic acid having an alicyclic skeleton at the 1-position include 1-cyclobutyl-1,1-difluoromethanesulfonic acid, 1-cyclopentyl-1,1-difluoromethanesulfonic acid, 1-cyclohexyl-1,1-difluoromethanesulfonic acid, 1-phenyl-1,1-difluoromethanesulfonic acid, 1-(4-trifluoromethylphenyl)-1,1-difluoromethanesulfonic acid, 1-(2,3-difluorophenyl)-1,1-difluoromethanesulfonic acid, 1-(2,4-difluorophenyl)-1,1-difluoromethanesulfonic acid, 1-(2,5-difluorophenyl)-1,1-difluoromethanesulfonic acid, 1-(2,6-difluorophenyl)-1,1-difluoromethanesulfonic acid, 1-(3,4-difluorophenyl)-1,1-difluoromethanesulfonic acid, 1-(3,5-difluorophenyl)-1,1-difluoromethanesulfonic acid, 1-(3,6-difluorophenyl)-1,1-difluoromethanesulfonic acid, 1-(2,3,4,5,6-pentafluorophenyl)-1,1-difluoromethanesulfonic acid, 1-(naphthalen-1-yl)-1,1-difluoromethanesulfonic acid, 1-(naphthalen-1-yl)-1,1-difluoromethanesulfonic acid, 1-(2-bicyclo[2.2.1]heptyl)-1,1-difluoromethanesulfonic acid, 1-(5-hydroxy-2-bicyclo[2.2.1]heptyl)-1,1-difluoromethanesulfonic acid, 1-(6-hydroxy-2-bicyclo[2.2.1]heptyl)-1,1-difluoromethanesulfonic acid, 1-(7,7-dimethyl-2-bicyclo[2.2.1]heptyl)-1,1-difluoromethanesulfonic acid, 1-tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl-1,1-difluoromethanesulfonic acid, 1-(9-hydroxytetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl)-1,1-difluoromethanesulfonic acid, 1-(10-hydroxytetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl)-1,1-difluoromethanesulfonic acid, 1-adamantan-1-yl-1,1-difluoromethanesulfonic acid, 1-adamantan-2-yl-1,1-difluoromethanesulfonic acid, 1-(3-hydroxyadamantan-1-yl)-1,1-difluoromethanesulfonic acid, 1-(3-hydroxyadamantan-1-yl)-1,1-difluoromethanesulfonic acid, and the like.

[0088]

In addition, examples of the 1,1-difluoromethanesulfonic acid having an alicyclic skeleton at the 2-position include 2-cyclobutyl-1,1-difluoroethanesulfonic acid, 2-cyclopentyl-1,1-difluoroethanesulfonic acid, 2-cyclohexyl-1,1-difluoroethanesulfonic acid, 2-phenyl-1,1-difluoroethanesulfonic acid,

2-(4-trifluoromethylphenyl)-1,1-difluoroethanesulfonic acid,
 2-(2,3-difluorophenyl)-1,1-difluoroethanesulfonic acid,
 2-(2,4-difluorophenyl)-1,1-difluoroethanesulfonic acid,
 2-(2,5-difluorophenyl)-1,1-difluoroethanesulfonic acid,
 2-(2,6-difluorophenyl)-1,1-difluoroethanesulfonic acid,
 2-(3,4-difluorophenyl)-1,1-difluoroethanesulfonic acid,
 2-(3,5-difluorophenyl)-1,1-difluoroethanesulfonic acid,
 2-(3,6-difluorophenyl)-1,1-difluoroethanesulfonic acid,
 2-(2,3,4,5,6-pentaluorophenyl)-1,1-difluoroethanesulfonic acid,
 2-(naphthalen-1-yl)-1,1-difluoroethanesulfonic acid,
 2-(naphthalen-2-yl)-1,1-difluoroethanesulfonic acid,
 2-(2-bicyclo[2.2.1]heptyl)-1,1-difluoroethanesulfonic acid,
 2-(5-hydroxy-2-bicyclo[2.2.1]heptyl)-1,1-difluoroethanesulfonic acid,
 2-(6-hydroxy-2-bicyclo[2.2.1]heptyl)-1,1-difluoroethanesulfonic acid,
 2-(7,7-dimethyl-2-bicyclo[2.2.1]heptyl)-1,1-difluoroethanesulfonic acid,
 2-tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl-1,1-difluoroethanesulfonic acid,
 2-(9-hydroxytetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl)-1,1-difluoroethanesulfonic acid,
 2-(10-hydroxytetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl)-1,1-difluoroethanesulfonic acid,
 2-adamantan-1-yl-1,1-difluoroethanesulfonic acid,
 2-adamantan-2-yl-1,1-difluoroethanesulfonic acid,
 2-(3-hydroxyadamantan-1-yl)-1,1-difluoroethanesulfonic acid,
 2-(3-hydroxyadamantan-2-yl)-1,1-difluoroethanesulfonic acid and the like.

[0089]

Specific examples of the acid represented by the above formula (VII) include linear, branched or cyclic alkyl sulfonic acid, aromatic sulfonic acid, 10-camphor sulfonic acid, an acid in which -SO₃-H group bonds to the substituent Rs when Rs is a cyclic hydrocarbon group having 3-20 carbon atoms, a cyclic fluorohydrocarbon group having 3-20 carbon atoms, or a derivative substituent of these groups, and the like.

Examples of the linear, branched or cyclic alkyl sulfonic acid include methanesulfonic acid, ethanesulfonic acid, n-propanesulfonic acid, n-butanesulfonic acid, i-butanesulfonic acid, sec-butanesulfonic acid, t-butanesulfonic acid, n-pentanesulfonic acid, n-hexanesulfonic acid, n-octanesulfonic acid, cyclopentanesulfonic acid, cyclohexanesulfonic acid and the like.

Additionally, examples of the aromatic sulfonic acid include benzenesulfonic acid, p-toluenesulfonic acid, benzylnesulfonic acid, α-naphthalenesulfonic acid, β-naphthalenesulfonic acid and the like.

[0090]

Specific examples of the acid represented by the above formula (VIII) include acetic acid, n-propionic acid, butyric acid, isobutyric acid, valeric acid, isovaleric acid, caproic acid, benzoic acid, salicylic acid, phthalic acid, terephthalic acid, α-naphthalenecarboxylic acid, β-naphthalenecarboxylic acid, cyclobutanecarboxylic acid, cyclopentanecarboxylic acid,

cyclohexanecarboxylic acid, 1,1-cyclobutanedicarboxylic acid, 1,2-cyclobutanedicarboxylic acid, 1,1-cyclopentanedicarboxylic acid, 1,2-cyclopentanedicarboxylic acid, 1,3-cyclopentanedicarboxylic acid, 1,1-cyclohexanedicarboxylic acid, 1,2-cyclohexanedicarboxylic acid, 1,3-cyclohexanedicarboxylic acid, 1,4-cyclohexanedicarboxylic acid, 2-norbornanecarboxylic acid, 2,3-norbornanedicarboxylic acid, norbornyl-2-acetic acid, 1-adamantanecarboxylic acid, 1-adamantaneacetic acid, 1,3-adamantanedicarboxylic acid, 1,3-adamantanedi-acetic acid, lithocholic acid, deoxycholic acid, chenodeoxycholic acid, cholic acid, an acid in which -COOH group bonds to the substituent R_c when R_c is a cyclic hydrocarbon group having 3-20 carbon atoms, a cyclic fluorohydrocarbon group having 3-20 carbon atoms, or a derivative substituent of these groups, and the like.

[0091]

Specific examples of the acid represented by the formula (IX) include N,N-bis(trifluoromethanesulfonyl)imidic acid, N,N-bis(pentafluoroethanesulfonyl)imidic acid, N,N-bis(heptafluoro-n-propanesulfonyl)imidic acid, N,N-bis(nonafluoro-n-butan-sulfonyl)imidic acid, N,N-bis(perfluoro-n-octanesulfonyl)imidic acid, N,N-bis(1,1,2,2-tetrafluoro-n-propanesulfonyl)imidic acid, N,N-bis(1,1,2,2-tetrafluoro-n-butan-sulfonyl)imidic acid, N,N-bis(1,1,2,2-tetrafluoro-n-octanesulfonyl)imidic acid, N-trifluoromethanesulfonyl-N-pentafluoroethanesulfonylimidic acid, N-trifluoromethanesulfonyl-N-heptafluoro-n-propanesulfonylimidic acid, N-trifluoromethanesulfonyl-N-nonafluoro-n-butan-sulfonylimidic acid, N-pentafluoroethanesulfonyl-N-heptafluoro-n-propanesulfonylimidic acid, N-pentafluoroethanesulfonyl-N-nonafluoro-n-butan-sulfonylimidic acid, N-heptafluoro-n-propanesulfonyl-N-nonafluoro-n-butan-sulfonylimidic acid, an acid in which -NH- group bonds to the substituent R_e when R_e is a cyclic hydrocarbon group having 3-20 carbon atoms, a cyclic fluorohydrocarbon group having 3-20 carbon atoms, or a derivative substituent of these group, and the like.

[0092]

2-2. Parent nucleus

Examples of the parent nucleus generating the acid represented by the formulas (V)-(IX) include an onium salt, a sulfoneimide compound, a sulfone compound, a sulfonate compound, a disulfonyldiazomethane compound, a disulfonylmethane compound, an oxime sulfonate compound, a hydrazine sulfonate compounds and the like.

[0093]

Examples of onium salt parent nuclei include an iodonium salt, a sulfonium salt (including a tetrahydrothiophenium salt), a phosphonium salt, a diazonium salt and a pyridinium salt.

Examples of the iodonium salt include diphenyliodonium salt, bis(4-t-butylphenyl)iodonium salt, di(p-tolyl)iodonium salt, di(3,4-dimethylphenyl)iodonium

salt, 4-nitrophenyl·phenyliodonium salt, di(3-nitrophenyl)iodonium salt, 4-methoxyphenyl·phenyliodonium salt, di(4-chlorophenyl)iodonium salt, di(4-trifluoromethylphenyl)iodonium salt, biphenyleneiodonium salt, di(2-naphthyl)iodonium salt, 2-chlorobiphenyleneiodonium salt and the like.

[0094]

Examples of the sulfonium salt include an aryl sulfonium salt such as triphenylsulfonium salt, 4-t-butylphenyl·diphenylsulfonium salt, 4-t-butoxyphenyl·diphenylsulfonium salt, 4-hydroxyphenyl·diphenylsulfonium salt, tri(4-methoxyphenyl)sulfonium salt, di(4-methoxyphenyl)·p-tolylsulfonium salt, phenyl·biphenylenesulfonium salt, (4-phenylthiophenyl)·diphenylsulfonium salt, and 4,4'-bis(diphenylsulfoniophenyl)sulfide salt; a (cyclo)alkylsulfonium salt such as dicyclohexylmethylsulfonium salt, dimethylcyclohexylsulfonium salt and tricyclohexylsulfonium salt; an 2-oxosulfonium salts such as cyclohexyl·2-oxocyclohexyl·methylsulfonium salt, dicyclohexyl·2-oxocyclohexylsulfonium salt, 2-oxocyclohexyldimethylsulfonium salt, bicyclo[2.2.1]hept-2-yl·methyl·2-(oxocyclohexyl)sulfonium salt, bicyclo[2.2.1]hept-2-yl·cyclohexyl·(2-oxocyclohexyl)sulfonium salt, 1-(2-naphthalen-1-yl-2-oxoethyl)tetrahydrothiophenium salt, 1-(2-naphthalen-2-yl-2-oxoethyl)tetrahydrothiophenium salt and 1-(2-oxo-n-butyl)tetrahydrothiophenium salt; a dialkyl·naphthalen-1-yl sulfonium salt such as 1-(naphthalen-1-yl)dimethylsulfonium salt, 1-(naphthalen-1-yl)diethylsulfonium salt, 1-(4-cyanonaphthalen-1-yl)dimethylsulfonium salt, 1-(4-cyanonaphthalen-1-yl)diethylsulfonium salt, 1-(4-nitronaphthalen-1-yl)dimethylsulfonium salt, 1-(4-nitronaphthalen-1-yl)diethylsulfonium salt, 1-(4-methylnaphthalen-1-yl)dimethylsulfonium salt, 1-(4-methylnaphthalen-1-yl)diethylsulfonium salt, 1-(4-hydroxynaphthalen-1-yl)tetrahydrothiophenium salt, 1-(4-hydroxynaphthalen-1-yl)dimethylsulfonium salt, 1-(4-hydroxynaphthalen-1-yl)diethylsulfonium salt and the like.

[0095]

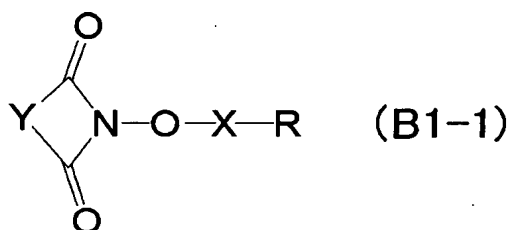
Examples of the arylthiophenium salt include 1-(4-methoxynaphthalen-1-yl)tetrahydrothiophenium salt, 1-(4-ethoxynaphthalen-1-yl)tetrahydrothiophenium salt, 1-(4-n-butoxynaphthalen-1-yl)tetrahydrothiophenium salt, 1-(4-methoxymethoxynaphthalen-1-yl)tetrahydrothiophenium salt, 1-(4-ethoxymethoxynaphthalen-1-yl)tetrahydrothiophenium salt, 1-[4-(1-methoxyethoxy)naphthalen-1-yl]tetrahydrothiophenium salt, 1-[4-(2-methoxyethoxy)naphthalen-1-yl]tetrahydrothiophenium salt, 1-(4-methoxycarbonyloxynaphthalen-1-yl)tetrahydrothiophenium salt, 1-(4-ethoxycarbonyloxynaphthalen-1-yl)tetrahydrothiophenium salt,

1-(4-n-propoxycarbonyloxynaphthalen-1-yl)tetrahydrothiophenium salt,
 1-(4-i-propoxycarbonyloxynaphthalen-1-yl)tetrahydrothiophenium salt,
 1-(4-n-butoxycarbonyloxynaphthalen-1-yl)tetrahydrothiophenium salt,
 1-(4-t-butoxycarbonyloxynaphthalen-1-yl)tetrahydrothiophenium salt,
 1-[4-(2-tetrahydrofuranyloxy)naphthalen-1-yl]tetrahydrothiophenium salt,
 1-[4-(2-tetrahydropyranyloxy)naphthalen-1-yl]tetrahydrothiophenium salt,
 1-(4-benzyloxynaphthalen-1-yl)tetrahydrothiophenium salt,
 4-(4-n-butoxynaphthalen-1-yl)-4-thioniatricyclo[5.2.1.0^{2,6}]decane salt,
 (4-ethoxynaphthalen-1-yl)-4-thioniatricyclo[5.2.1.0^{2,6}]decane salt,
 1-[4-(bicyclo[2.2.1]hept-2-yl)oxynaphthalen-1-yl]tetrahydrothiophenium salt,
 1-(3,5-dimethyl-4-hydroxyphenyl)tetrahydrothiophenium salt,
 1-(3,5-dimethyl-4-ethoxyphenyl)tetrahydrothiophenium salt,
 1-(3,5-dimethyl-4-butoxyphenyl)tetrahydrothiophenium salt and the like.

[0096]

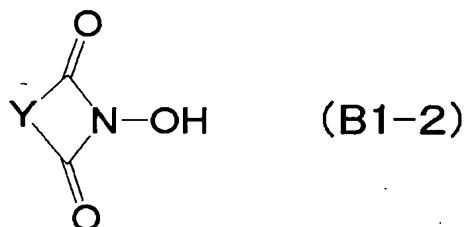
The sulfonimide compound is not particularly limited, but examples include a compound represented by the general formula (B1-1) below. This compound is formed by a sulfonyloxy bond or ester bond of a parent nucleus represented by the general formula (B1-2) below with an acid generated.

[0097]



(In the formula, R represents a skeleton of an acid by the above general formula (V), (VI), (VII), (VIII) or (IX), X represents a bonding group derived from the acid generated, and Y represents a divalent organic group.)

[0098]



(Y represents a divalent organic group.)

[0099]

In the general formula (B1-1) above, when the acid generated is a sulfonic acid, X represents a sulfonyl group, and when the acid generated is a carboxylic acid, X represents a

carbonyl group. Additionally, when the acid generated is an imidic acid, X represents N-R' (R' represents a sulfonyl group-containing organic group or a carbonyl group-containing organic group.) and the groups R and R' depend on the structure of the generated acid.

Further, Y in the general formulas (B1-1) and (B1-2) above is not particularly limited, but examples include an alkylene group such as methylene group, ethylene group, n-propylene group, isopropylene group, n-butylene group and isobutylene group; a divalent organic group derived from a cycloalkane such as cyclopentane, cyclohexane, cycloheptane, cyclooctane and bicyclo[2.2.1]heptane or an organic alicyclic structure; and the like.

[0100]

Examples of the mother nucleus represented by the general formula (B1-2) forming a sulfonimide compound include N-hydroxysuccinimide, N-hydroxydiphenylmaleimide, N-hydroxybicyclo[2.2.1]hept-5-ene-2,3-dicarboxyimide, N-hydroxy-7-oxabicyclo[2.2.1]hept-5-ene-2,3-dicarboxyimide, N-hydroxybicyclo[2.2.1]heptane-5,6-oxy-2,3-dicarboxyimide, N-hydroxynaphthylimide, N-hydroxyphthalimide and the like.

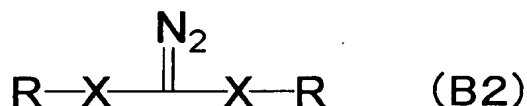
[0101]

Examples of the above sulfone compound include a β -ketosulfone, a β -sulfonylsulfone, an α -diazo compound of these compounds and the like.

Additionally, examples of the above sulfonate compound include an alkyl sulfonate, a haloalkyl sulfonate, an aryl sulfonate, an imino sulfonate and the like.

[0102]

The above disulfonyl diazomethane compound is not particularly limited, but examples include the compound represented by the general formula (B2) below.



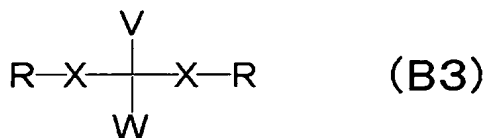
(In the formula, R represents a skeleton of an acid by the general formula (V), (VI), (VII), (VIII) and (IX) above, and X represents a bonding group derived from the acid generated.)

[0103]

In the general formula (B2) above, when the acid generated is a sulfonic acid, X represents a sulfonyl group, and when the acid generated is a carboxylic acid, X represents a carbonyl group. Additionally, when the acid generated is an imidic acid, X represents N-R' (R' represents a sulfonyl group-containing organic group or a carbonyl group-containing organic group.) and the groups R and R' depend on the structure of the generated acid.

[0104]

The above disulfonylmethane compound is not particularly limited, but examples include the compound represented by the general formula (B3) below.



(In the formula, R represents a skeleton of an acid by the general formula (V), (VI), (VII), (VIII) and (IX) above, X represents a bonding group derived from the acid generated, and V and W individually represent an aryl group, in combination form a monocyclic or polycyclic ring containing at least one unsaturated bond, or in combination form a group represented by the general formula (B4) below.)

[0105]

In the general formula (B3) above, when the acid generated is a sulfonic acid, X represents a sulfonyl group, and when the acid generated is a carboxylic acid, X represents a carbonyl group. Additionally, when the acid generated is an imidic acid, X represents N-R' (R' represents a sulfonyl group-containing organic group or a carbonyl group-containing organic group.) and the groups R and R' depend on the structure of the generated acid.

[0106]

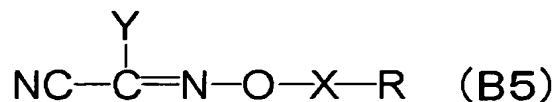


(In the formula, V' and W' may be either the same or different, plural V's and W's may be either the same or different, and each of them represents hydrogen atom, a halogen atom, an alkyl group, a cycloalkyl group, an aryl group or an aralkyl group, or V' and W' each bonded to the same or different carbon atom bond to form a monocyclic carbon structure, and k is an integer of 2-10.)

[0107]

The above oxime sulfonate compound is not particularly limited, but examples include the compound represented by the general formulas (B5) and (B6) below.

[0108]



(In the formula, R represents a skeleton of an acid by the general formula (V), (VI), (VII),

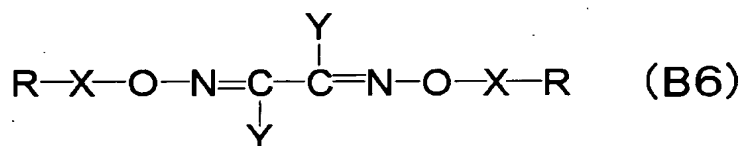
(VIII) or (IX) above, X represents a bonding group derived from the acid generated, and Y represents a monovalent organic group.)

[0109]

In the general formula (B5) above, when the acid generated is a sulfonic acid, X represents a sulfonyl group, and when the acid generated is a carboxylic acid, X represents a carbonyl group. Additionally, when the acid generated is an imidic acid, X represents N-R' (R' represents a sulfonyl group-containing organic group or a carbonyl group-containing organic group.) and the groups R and R' depend on the structure of the generated acid.

Examples of Y in the general formula (B5) above include methyl group, ethyl group, n-propyl group, phenyl group, tosyl group and the like.

[0110]



(In the formula, Rs individually represent a skeleton of an acid by the general formula (V), (VI), (VII), (VIII) or (IX) above, X represents a bonding group derived from the acid generated, and Ys represent individually a monovalent organic group.)

[0111]

In the general formula (B6) above, when the acid generated is a sulfonic acid, X represents a sulfonyl group, and when the acid generated is a carboxylic acid, X represents a carbonyl group. Additionally, when the acid generated is an imidic acid, X represents N-R' (R' represents a sulfonyl group-containing organic group or a carbonyl group-containing organic group.) and the groups R and R' depend on the structure of the generated acid.

Examples of Y in the general formula (B6) above include methyl group, ethyl group, n-propyl group, phenyl group, tosyl group and the like.

[0112]

Examples of the hydrazine sulfonate compound include bis(benzene)sulfonylhydrazine, bis(p-toluene)sulfonylhydrazine, bis(trifluoromethane)sulfonylhydrazine, bis(nonafluoro-n-butane)sulfonylhydrazine, bis(n-propane)sulfonylhydrazine, benzenesulfonylhydrazine, p-toluenesulfonylhydrazine, trifluoromethanesulfonylhydrazine, nonafluoro-n-butanedisulfonylhydrazine, n-propanedisulfonylhydrazine, trifluoromethanesulfonyl-p-toluenedisulfonylhydrazine and the like.

[0113]

2-3. Example of acid generator [B]

Examples of the acid generator [B] comprising the above acid generated and the above parent nucleus include diphenyliodonium trifluoromethanesulfonate, diphenyliodonium nonafluoro-n-butanedisulfonate, diphenyliodonium

perfluoro-n-octanesulfonate, diphenyliodonium
 2-bicyclo[2.2.1]hept-2-yl-1,1,2,2-tetrafluoroethanesulfonate, diphenyliodonium
 1,1,2,2-tetrafluoro-2-(5(6)-hydroxybicyclo[2.2.1]hept-2-yl) ethanesulfonate,
 diphenyliodonium
 2-tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl-1,1,2,2-tetrafluoroethanesulfonate,
 diphenyliodonium 2-(9(10)-hydroxy-3-tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl)-1,1,2,2-
 tetrafluoroethanesulfonate, diphenyliodonium N,N-bis(trifluoromethanesulfonyl)imide,
 diphenyliodonium N,N-bis(pentafluoroethanesulfonyl)imide, diphenyliodonium
 N,N-bis(heptafluoro-n-propanesulfonyl)imide, diphenyliodonium
 N,N-bis(nonafluoro-n-buthanesulfonyl)imide, diphenyliodonium benzenesulfonate,
 diphenyliodonium 4-trifluorobenzenesulfonate, diphenyliodonium
 2,4-difluorobenzenesulfonate, diphenyliodonium 2,3,4,5,6-pentafluorobenzenesulfonate,
 diphenyliodonium camphorsulfonate,

[0114]

bis(4-t-butylphenyl)iodoniumtrifluoromethane sulfonate,
 bis(4-t-butylphenyl)iodonium nonafluoro-n-buthanesulfonate, bis(4-t-butylphenyl)iodonium
 perfluoro-n-octanesulfonate, bis(4-t-butylphenyl)iodonium
 2-bicyclo[2.2.1]hept-2-yl-1,1,2,2-tetrafluoroethanesulfonate, bis(4-t-butylphenyl)iodonium
 1,1,2,2-tetrafluoro-2-(5(6)-hydroxybicyclo[2.2.1]hept-2-yl)ethanesulfonate,
 bis(4-t-butylphenyl)iodonium
 2-tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl-1,1,2,2-tetrafluoroethanesulfonate,
 bis(4-t-butylphenyl)iodonium
 2-(9(10)-hydroxytetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl)-1,1,2,2- tetrafluoroethanesulfonate,
 bis(4-t-butylphenyl)iodonium N,N-bis(trifluoromethanesulfonyl)imide,
 bis(4-t-butylphenyl)iodonium N,N-bis(pentafluoroethanesulfonyl)imide,
 bis(4-t-butylphenyl)iodonium N,N-bis(heptafluoro-n-propanesulfonyl)imide,
 bis(4-t-butylphenyl)iodonium N,N-bis(nonafluoro-n-buthanesulfonyl)imide,
 bis(4-t-butylphenyl)iodonium benzenesulfonate, bis(4-t-butylphenyl)iodonium
 4-trifluorobenzenesulfonate, bis(4-t-butylphenyl)iodonium 2,4-difluorobenzenesulfonate,
 bis(4-t-butylphenyl)iodonium 2,3,4,5,6-pentafluorobenzenesulfonate,
 bis(4-t-butylphenyl)iodonium camphorsulfonate,

[0115]

triphenylsulfonium trifluoromethanesulfonate, triphenylsulfonium
 nonafluoro-n-buthanesulfonate, triphenylsulfonium perfluoro-n-octanesulfonate,
 triphenylsulfonium 2-bicyclo[2.2.1]hept-2-yl-1,1,2,2-tetrafluoroethanesulfonate,
 triphenylsulfonium 1,1,2,2-tetrafluoro-2-(5(6)-hydroxybicyclo[2.2.1]hept-2-yl)
 ethanesulfonate, triphenylsulfonium
 2-tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl-1,1,2,2-tetrafluoroethanesulfonate,
 triphenyliodonium
 2-(9(10)-hydroxytetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl)-1,1,2,2-tetrafluoroethanesulfonate,
 triphenylsulfonium N,N-bis(trifluoromethanesulfonyl)imide, triphenylsulfonium

N,N-bis(pentafluoroethanesulfonyl)imide, triphenylsulfonium
 N,N-bis(heptafluoro-n-propanesulfonyl)imide, triphenylsulfonium
 N,N-bis(nonafluoro-n-buthanesulfonyl)imide, triphenylsulfonium benzenesulfonate,
 triphenylsulfonium 4-trifluoromethylbenzenesulfonate, triphenylsulfonium
 2,4-difluorobenzenesulfonate, triphenylsulfonium 2,3,4,5,6-pentafluorobenzenesulfonate,
 triphenylsulfonium camphorsulfonate,

[0116]

bicyclo[2.2.1]heptan-2-yl•cyclohexyl•(2-oxocyclohexyl)sulfonium
 trifluoromethanesulfonate, bicyclo[2.2.1]hept-2-yl•cyclohexyl•(2-oxocyclohexyl)sulfonium
 nonafluoro-n-buthanesulfonate,
 bicyclo[2.2.1]hept-2-yl•cyclohexyl•(2-oxocyclohexyl)sulfonium perfluoro-n-octanesulfonate,
 bicyclo[2.2.1]hept-2-yl•cyclohexyl•(2-oxocyclohexyl)sulfonium
 2-bicyclo[2.2.1]heptan-2-yl-1,1,2,2-tetrafluoroethanesulfonate,
 bicyclo[2.2.1]hept-2-yl•cyclohexyl•(2-oxocyclohexyl)sulfonium
 1,1,2,2-tetrafluoro-2-(5(6)-hydroxybicyclo[2.2.1]hept-2-yl)ethanesulfonate,
 bicyclo[2.2.1]hept-2-yl•cyclohexyl•(2-oxocyclohexyl)sulfonium 2-tetracyclo
 [6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl-1,1,2,2-tetrafluoroethanesulfonate,
 bicyclo[2.2.1]hept-2-yl•cyclohexyl•(2-oxocyclohexyl)sulfonium 2-(9(10)-hydroxytetracyclo
 [6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl)-1,1,2,2-tetrafluoroethanesulfonate,
 bicyclo[2.2.1]hept-2-yl•cyclohexyl•(2-oxocyclohexyl)sulfonium
 N,N-bis(trifluoromethanesulfonyl)imide,
 bicyclo[2.2.1]hept-2-yl•cyclohexyl•(2-oxocyclohexyl)sulfonium
 N,N-bis(pentafluoroethanesulfonyl)imide,
 bicyclo[2.2.1]heptan-2-yl•cyclohexyl•(2-oxocyclohexyl)sulfonium
 N,N-bis(heptafluoro-n-propanesulfonyl)imide,
 bicyclo[2.2.1]hept-2-yl•cyclohexyl•(2-oxocyclohexyl)sulfonium
 N,N-bis(nonafluoro-n-buthanesulfonyl)imide,
 bicyclo[2.2.1]hept-2-yl•cyclohexyl•(2-oxocyclohexyl)sulfonium benzenesulfonate,
 bicyclo[2.2.1]hept-2-yl•cyclohexyl•(2-oxocyclohexyl)sulfonium 4-trifluorobenzenesulfonate,
 bicyclo[2.2.1]hept-2-yl•cyclohexyl•(2-oxocyclohexyl)sulfonium
 2,4-trifluorobenzenesulfonate,
 bicyclo[2.2.1]hept-2-yl•cyclohexyl•(2-oxocyclohexyl)sulfonium
 2,3,4,5,6-pentafluorobenzenesulfonate,
 bicyclo[2.2.1]hept-2-yl•cyclohexyl•(2-oxocyclohexyl)sulfonium camphorsulfonate,

[0117]

1-(2-naphthalen-1-yl-2-oxoethyl)tetrahydrothiophenium trifluoromethanesulfonate,
 1-(2-naphthalen-1-yl-2-oxoethyl)tetrahydrothiophenium nonafluoro-n-buthanesulfonate,
 1-(2-naphthalen-1-yl-2-oxoethyl)tetrahydrothiophenium perfluoro-n-octanesulfonate,
 1-(2-naphthalen-1-yl-2-oxoethyl)tetrahydrothiophenium
 2-bicyclo[2.2.1]hept-2-yl-1,1,2,2-tetrafluoroethanesulfonate,
 1-(2-naphthalen-1-yl-2-oxoethyl)tetrahydrothiophenium

1,1,2,2-tetrafluoro-2-(5(6)-hydroxybicyclo[2.2.1]hept-2-yl)ethanesulfonate,
 1-(2-naphthalen-1-yl-2-oxoethyl)tetrahydrothiophenium
 2-tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl-1,1,2,2-tetrafluoroethanesulfonate,
 1-(2-naphthalen-1-yl-2-oxoethyl)tetrahydrothiophenium
 2-(8(9)-hydroxytetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl)-1,1,2,2- tetrafluoroethanesulfonate,
 1-(2-naphthalen-1-yl-2-oxoethyl)tetrahydrothiophenium
 N,N-bis(trifluoromethanesulfonyl)imide,
 1-(2-naphthalen-1-yl-2-oxoethyl)tetrahydrothiophenium
 N,N-bis(pentafluoroethanesulfonyl)imide,
 1-(2-naphthalen-1-yl-2-oxoethyl)tetrahydrothiophenium
 N,N-bis(heptafluoro-n-propanesulfonyl)imide,
 1-(2-naphthalen-1-yl-2-oxoethyl)tetrahydrothiophenium
 N,N-bis(nonafluoro-n-butanessulfonyl)imide,
 1-(2-(naphthalen-1-yl-2-oxoethyl)tetrahydrothiophenium benzenesulfonate,
 1-(2-naphthalen-1-yl-2-oxoethyl)tetrahydrothiophenium 4-trifluoromethylbenzenesulfonate,
 1-(2-naphthalen-1-yl-2-oxoethyl)tetrahydrothiophenium 2,4-difluorobenzenesulfonate,
 1-(2-naphthalen-1-yl-2-oxoethyl)tetrahydrothiophenium
 2,3,4,5,6-pentafluorobenzenesulfonate,
 1-(2-naphthalen-1-yl-2-oxoethyl)tetrahydrothiophenium camphorsulfonate,
 [0118]
 1-(4-hydroxynaphthalen-1-yl)tetrahydrothiophenium trifluoromethanesulfonate,
 diphenyliodonium nonafluoro-n-butanessulfonate,
 1-(4-hydroxynaphthalen-1-yl)tetrahydrothiophenium perfluoro-n-octanesulfonate,
 1-(4-hydroxynaphthalen-1-yl)tetrahydrothiophenium
 2-bicyclo[2.2.1]heptan-2-yl-1,1,2,2-tetrafluoroethanesulfonate,
 1-(4-hydroxynaphthalen-1-yl)tetrahydrothiophenium
 1,1,2,2-tetrafluoro-2-(5(6)-hydroxybicyclo[2.2.1]hept-2-yl)ethanesulfonate,
 1-(4-hydroxynaphthalen-1-yl)tetrahydrothiophenium
 2-tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl-1,1,2,2-tetrafluoroethanesulfonate,
 1-(4-hydroxynaphthalen-1-yl)tetrahydrothiophenium
 2-(9(10)-hydroxytetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl)-1,1,2,2- tetrafluoroethanesulfonate,
 1-(4-hydroxynaphthalen-1-yl)tetrahydrothiophenium
 N,N-bis(trifluoromethanesulfonyl)imide,
 1-(4-hydroxynaphthalen-1-yl)tetrahydrothiophenium
 N,N-bis(pentafluoroethanesulfonyl)imide,
 1-(4-hydroxynaphthalen-1-yl)tetrahydrothiophenium
 N,N-bis(heptafluoro-n-propanesulfonyl)imide,
 1-(4-hydroxynaphthalen-1-yl)tetrahydrothiophenium
 N,N-bis(nonafluoro-n-butanessulfonyl)imide,
 1-(4-hydroxynaphthalen-1-yl)tetrahydrothiophenium benzenesulfonate, diphenyliodonium
 4-trifluorobenzenesulfonate, 1-(4-hydroxynaphthalen-1-yl)tetrahydrothiophenium

2,4-difluorobenzenesulfonate, 1-(4-hydroxynaphthalen-1-yl)tetrahydrothiophenium
2,3,4,5,6-pentafluorobenzenesulfonate, 1-(4-hydroxynaphthalen-1-yl)tetrahydrothiophenium
camphorsulfonate,

[0119]

1-(4-n-butoxynaphthalen-1-yl)tetrahydrothiophenium trifluoromethanesulfonate,
1-(4-n-butoxynaphthalen-1-yl)tetrahydrothiophenium nonafluoro-n-butanesulfonate,
1-(4-n-butoxynaphthalen-1-yl)tetrahydrothiophenium perfluoro-n-octanesulfonate,
1-(4-n-butoxynaphthalen-1-yl)tetrahydrothiophenium
2-bicyclo[2.2.1]heptan-2-yl-1,1,2,2-tetrafluoroethanesulfonate,
1-(4-n-butoxynaphthalen-1-yl)tetrahydrothiophenium
1,1,2,2-tetrafluoro-2-(5(6)-hydroxybicyclo[2.2.1]hept-2-yl)ethanesulfonate,
1-(4-n-butoxynaphthalen-1-yl)tetrahydrothiophenium
2-tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl-1,1,2,2-tetrafluoroethanesulfonate,
1-(4-n-butoxynaphthalen-1-yl)tetrahydrothiophenium
2-(9(10)-hydroxytetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl)-1,1,2,2-tetrafluoroethanesulfonate,
1-(4-n-butoxynaphthalen-1-yl)tetrahydrothiophenium
N,N-bis(trifluoromethanesulfonyl)imide,
1-(4-n-butoxynaphthalen-1-yl)tetrahydrothiophenium
N,N-bis(pentafluoroethanesulfonyl)imide,
1-(4-n-butoxynaphthalen-1-yl)tetrahydrothiophenium
N,N-bis(heptafluoro-n-propanesulfonyl)imide,
1-(4-n-butoxynaphthalen-1-yl)tetrahydrothiophenium
N,N-bis(nonafluoro-n-butanesulfonyl)imide,
1-(4-n-butoxynaphthalen-1-yl)tetrahydrothiophenium benzenesulfonate,
1-(4-n-butoxynaphthalen-1-yl)tetrahydrothiophenium 4-trifluorobenzenesulfonate,
1-(4-n-butoxynaphthalen-1-yl)tetrahydrothiophenium 2,4-difluorobenzenesulfonate,
1-(4-n-butoxynaphthalen-1-yl)tetrahydrothiophenium 2,3,4,5,6-pentafluorobenzenesulfonate,
1-(4-n-butoxynaphthalen-1-yl)tetrahydrothiophenium camphorsulfonate,

[0120]

(4-n-butoxynaphthalen-1-yl)-4-thioniatricyclo[5.2.1.0^{2,6}]decane
trifluoromethanesulfonate, (4-n-butoxynaphthalen-1-yl)-4-thioniatricyclo[5.2.1.0^{2,6}]decane
nonafluoro-n-butanesulfonate,
(4-n-butoxynaphthalen-1-yl)-4-thioniatricyclo[5.2.1.0^{2,6}]decane perfluoro-n-octanesulfonate,
(4-n-butoxynaphthalen-1-yl)-4-thioniatricyclo[5.2.1.0^{2,6}]decane
2-bicyclo[2.2.1]hept-2-yl-1,1,2,2-tetrafluoroethanesulfonate,
(4-n-butoxynaphthalen-1-yl)-4-thioniatricyclo[5.2.1.0^{2,6}]decane
1,1,2,2-tetrafluoro-2-(5(6)-hydroxybicyclo[2.2.1]hept-2-yl)ethanesulfonate,
(4-n-butoxynaphthalen-1-yl)-4-thioniatricyclo[5.2.1.0^{2,6}]decane
2-tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl-1,1,2,2-tetrafluoroethanesulfonate,
(4-n-butoxynaphthalen-1-yl)-4-thioniatricyclo[5.2.1.0^{2,6}]decane
2-(9(10)-hydroxytetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl)-1,1,2,2-tetrafluoroethanesulfonate,

(4-n-butoxynaphthalen-1-yl)-4-thioniatricyclo[5.2.1.0^{2,6}]decane
 N,N-bis(trifluoromethanesulfonyl)imide,
 (4-n-butoxynaphthalen-1-yl)-4-thioniatricyclo[5.2.1.0^{2,6}]decane
 N,N-bis(pentafluoroethanesulfonyl)imide,
 (4-n-butoxynaphthalen-1-yl)-4-thioniatricyclo[5.2.1.0^{2,6}]decane
 N,N-bis(heptafluoro-n-propanesulfonyl)imide,
 (4-n-butoxynaphthalen-1-yl)-4-thioniatricyclo[5.2.1.0^{2,6}]decane
 N,N-bis(nonafluoro-n-buthanesulfonyl)imide, diphenyliodonium benzenesulfonate,
 (4-n-butoxynaphthalen-1-yl)-4-thioniatricyclo[5.2.1.0^{2,6}]decane 4-trifluorobenzenesulfonate,
 (4-n-butoxynaphthalen-1-yl)-4-thioniatricyclo[5.2.1.0^{2,6}]decane
 2,4-difluorobenzenesulfonate,
 (4-n-butoxynaphthalen-1-yl)-4-thioniatricyclo[5.2.1.0^{2,6}]decane
 2,3,4,5,6-pentafluorobenzenesulfonate,
 (4-n-butoxynaphthalen-1-yl)-4-thioniatricyclo[5.2.1.0^{2,6}]decane camphorsulfonate,

[0121]

1-(3,5-dimethyl-4-hydroxyphenyl)tetrahydrothiophenium trifluoromethanesulfonate,
 1-(3,5-dimethyl-4-hydroxyphenyl)tetrahydrothiophenium nonafluoro-n-buthanesulfonate,
 1-(3,5-dimethyl-4-hydroxyphenyl)tetrahydrothiophenium perfluoro-n-octanesulfonate,
 1-(3,5-dimethyl-4-hydroxyphenyl)tetrahydrothiophenium
 2-(bicyclo[2.2.1]hept-2-yl)-1,1,2,2-tetrafluoroethanesulfonate,
 1-(3,5-dimethyl-4-hydroxyphenyl)tetrahydrothiophenium
 1,1,2,2-tetrafluoro-2-(5(6)-hydroxybicyclo[2.2.1]hept-2-yl)ethanesulfonate,
 1-(3,5-dimethyl-4-hydroxyphenyl)tetrahydrothiophenium
 2-tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl-1,1,2,2-tetrafluoroethanesulfonate,
 1-(3,5-dimethyl-4-hydroxyphenyl)tetrahydrothiophenium
 2-(9(10)-hydroxytetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl)-1,1,2,2-tetrafluoroethanesulfonate,
 1-(3,5-dimethyl-4-hydroxyphenyl)tetrahydrothiophenium
 N,N-bis(trifluoromethanesulfonyl)imide,
 1-(3,5-dimethyl-4-hydroxyphenyl)tetrahydrothiophenium
 N,N-bis(pentafluoroethanesulfonyl)imide,
 1-(3,5-dimethyl-4-hydroxyphenyl)tetrahydrothiophenium
 N,N-bis(heptafluoro-n-propanesulfonyl)imide,
 1-(3,5-dimethyl-4-hydroxyphenyl)tetrahydrothiophenium
 N,N-bis(nonafluoro-n-buthanesulfonyl)imide,
 1-(3,5-dimethyl-4-hydroxyphenyl)tetrahydrothiophenium benzenesulfonate,
 1-(3,5-dimethyl-4-hydroxyphenyl)tetrahydrothiophenium 4-trifluorobenzenesulfonate,
 1-(3,5-dimethyl-4-hydroxyphenyl)tetrahydrothiophenium 2,4-difluorobenzenesulfonate,
 1-(3,5-dimethyl-4-hydroxyphenyl)tetrahydrothiophenium
 2,3,4,5,6-pentafluorobenzenesulfonate,
 1-(3,5-dimethyl-4-hydroxyphenyl)tetrahydrothiophenium camphorsulfonate,

[0122]

1-(3,5-dimethyl-4-butoxyphenyl)tetrahydrothiophenium trifluoromethanesulfonate,
 1-(3,5-dimethyl-4-butoxyphenyl)tetrahydrothiophenium nonafluoro-n-butanesulfonate,
 1-(3,5-dimethyl-4-butoxyphenyl)tetrahydrothiophenium perfluoro-n-octanesulfonate,
 1-(3,5-dimethyl-4-butoxyphenyl)tetrahydrothiophenium
 2-(bicyclo[2.2.1]hept-2-yl)-1,1,2,2-tetrafluoroethanesulfonate,
 1-(3,5-dimethyl-4-butoxyphenyl)tetrahydrothiophenium
 1,1,2,2-tetrafluoro-2-(5(6)-hydroxybicyclo[2.2.1]hept-2-yl)ethanesulfonate,
 1-(3,5-dimethyl-4-butoxyphenyl)tetrahydrothiophenium
 2-tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl-1,1,2,2-tetrafluoroethanesulfonate,
 1-(3,5-dimethyl-4-butoxyphenyl)tetrahydrothiophenium
 2-(9(10)-hydroxytetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl)-1,1,2,2-tetrafluoroethanesulfonate,
 1-(3,5-dimethyl-4-butoxyphenyl)tetrahydrothiophenium
 N,N-bis(trifluoromethanesulfonyl)imide,
 1-(3,5-dimethyl-4-butoxyphenyl)tetrahydrothiophenium
 N,N-bis(pentafluoroethanesulfonyl)imide,
 1-(3,5-dimethyl-4-butoxyphenyl)tetrahydrothiophenium
 N,N-bis(heptafluoro-n-propanesulfonyl)imide,
 1-(3,5-dimethyl-4-butoxyphenyl)tetrahydrothiophenium
 N,N-bis(nonafluoro-n-butanesulfonyl)imide,
 1-(3,5-dimethyl-4-butoxyphenyl)tetrahydrothiophenium benzenesulfonate,
 1-(3,5-dimethyl-4-butoxyphenyl)tetrahydrothiophenium 4-trifluorobenzenesulfonate,
 1-(3,5-dimethyl-4-butoxyphenyl)tetrahydrothiophenium 2,4-difluorobenzenesulfonate,
 1-(3,5-dimethyl-4-butoxyphenyl)tetrahydrothiophenium
 2,3,4,5,6-pentafluorobenzenesulfonate,
 1-(3,5-dimethyl-4-butoxyphenyl)tetrahydrothiophenium camphorsulfonate,

[0123]

N-(trifluoromethanesulfonyloxy)succinimide,
 N-(nonafluoro-n-butanesulfonyloxy)succinimide,
 N-(perfluoro-n-octanesulfonyloxy)succinimide,
 N-(2-bicyclo[2.2.1]hept-2-yl)-1,1,2,2-tetrafluoroethanesulfonyloxy)succinimide,
 N-(1,1,2,2-tetrafluoro-2-(5(6)-hydroxybicyclo[2.2.1]hept-2-yl)
 ethanesulfonyloxy)succinimide,
 N-(2-tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl)-1,1,2,2-tetrafluoroethanesulfonyloxy)
 succinimide,
 N-(2-(9(10)-hydroxytetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl)-1,1,2,2-tetrafluoroethanesulfonyl
 oxy)succinimide, N-(benzenesulfonyloxy)succinimide,
 N-(4-trifluoromethylbenzenesulfonyloxy)succinimide,
 N-(2,4-difluorobenzenesulfonyloxy)succinimide,
 N-(2,3,4,5,6-pentafluorobenzenesulfonyloxy)succinimide,
 N-(camphorsulfonyloxy)succinimide,

[0124]

N-(trifluoromethanesulfonyloxy)bicyclo[2.2.1]hept-5-ene-2,3-dicarboxyimide,
 N-(nonafluoro-n-butanesulfonyloxy)bicyclo[2.2.1]hept-5-ene-2,3-dicarboxyimide,
 N-(perfluoro-n-octanesulfonyloxy)bicyclo[2.2.1]hept-5-ene-2,3-dicarboxyimide,
 N-(2-bicyclo[2.2.1]hept-2-yl-1,1,2,2-tetrafluoroethanesulfonyloxy)bicyclo[2.2.1]hept-5-ene-2,3-dicarboxyimide,
 N-(1,1,2,2-tetrafluoro-2-(5(6)-hydroxybicyclo[2.2.1]hept-2-yl)ethanesulfonyloxy)bicyclo[2.2.1]hept-5-ene-2,3-dicarboxyimide,
 N-(2-tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl-1,1,2,2-tetrafluoroethanesulfonyloxy)bicyclo[2.2.1]hept-5-ene-2,3-dicarboxyimide,
 N-(2-(9(10)-hydroxytetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl)-1,1,2,2-tetrafluoroethanesulfonyloxy)bicyclo[2.2.1]hept-5-ene-2,3-dicarboxyimide,
 N-(benzenesulfonyloxy)bicyclo[2.2.1]hept-5-ene-2,3-dicarboxyimide,
 N-(4-trifluoromethylbenzenesulfonyloxy)bicyclo[2.2.1]hept-5-ene-2,3-dicarboxyimide,
 N-(2,4-difluorobenzenesulfonyloxy)bicyclo[2.2.1]hept-5-ene-2,3-dicarboxyimide,
 N-(2,3,4,5,6-pentafluorobenzenesulfonyloxy)bicyclo[2.2.1]hept-5-ene-2,3-dicarboxyimide,
 N-(camphorsulfonyloxy)bicyclo[2.2.1]hept-5-ene-2,3-dicarboxyimide,

[0125]

N-(trifluoromethanesulfonyloxy)-7-oxabicyclo[2.2.1]hept-5-ene-2,3-dicarboxyimide, N-(nonafluoro-n-butanesulfonyloxy)-7-oxabicyclo[2.2.1]hept-5-ene-2,3-dicarboxyimide, N-(perfluoro-n-octanesulfonyloxy)-7-oxabicyclo[2.2.1]hept-5-ene-2,3-dicarboxyimide, N-(2-bicyclo[2.2.1]hept-2-yl-1,1,2,2-tetrafluoroethanesulfonyloxy)-7-oxabicyclo[2.2.1]hept-5-ene-2,3-dicarboxyimide,
 N-(1,1,2,2-tetrafluoro-2-(5(6)-hydroxybicyclo[2.2.1]hept-2-yl)ethanesulfonyloxy)-7-bicyclo[2.2.1]hept-5-ene-2,3-dicarboxyimide,
 N-(2-tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl-1,1,2,2-tetrafluoroethanesulfonyloxy)-7-oxabicyclo[2.2.1]hept-5-ene-2,3-dicarboxyimide,
 N-(2-(9(10)-hydroxytetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl)-1,1,2,2-tetrafluoroethanesulfonyloxy)-7-oxabicyclo[2.2.1]hept-5-ene-2,3-dicarboxyimide,
 N-(benzenesulfonyloxy)-7-oxabicyclo[2.2.1]hept-5-ene-2,3-dicarboxyimide,
 N-(4-trifluoromethylbenzenesulfonyloxy)-7-oxabicyclo[2.2.1]hept-5-ene-2,3-dicarboxyimide, N-(2,4-difluorobenzenesulfonyloxy)-7-oxabicyclo[2.2.1]hept-5-ene-2,3-dicarboxyimide, N-(2,3,4,5,6-pentafluorobenzenesulfonyloxy)-7-oxabicyclo[2.2.1]hept-5-ene-2,3-dicarboxyimide, N-(camphorsulfonyloxy)-7-oxabicyclo[2.2.1]hept-5-ene-2,3-dicarboxyimide and the like.

[0126]

Among these, diphenyliodonium trifluoromethanesulfonate, diphenyliodonium nonafluoro-n-butanesulfonate, diphenyliodonium perfluoro-n-octanesulfonate, diphenyliodonium 2-bicyclo[2.2.1]hept-2-yl-1,1,2,2-tetrafluoroethanesulfonate, diphenyliodonium 2-tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl-1,1,2,2-tetrafluoroethanesulfonate, diphenyliodonium N,N-bis(nonafluoro-n-butanesulfonyl)imidate, diphenyliodonium

camphorsulfonate, bis(4-t-butylphenyl)iodoniumtrifluoromethane sulfonate,
 bis(4-t-butylphenyl)iodonium nonafluoro-n-butanesulfonate, bis(4-t-butylphenyl)iodonium
 perfluoro-n-octanesulfonate, bis(4-t-butylphenyl)iodonium
 2-bicyclo[2.2.1]hept-2-yl-1,1,2,2-tetrafluoroethanesulfonate, bis(4-t-butylphenyl)iodonium
 2-tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl-1,1,2,2-tetrafluoroethanesulfonate,
 bis(4-t-butylphenyl)iodonium N,N-bis(nonafluoro-n-butanesulfonyl)imide,
 bis(4-t-butylphenyl)iodonium camphorsulfonate, triphenylsulfonium
 trifluoromethanesulfonate, triphenylsulfonium nonafluoro-n-butanesulfonate,
 triphenylsulfonium perfluoro-n-octanesulfonate, triphenyliodonium
 2-bicyclo[2.2.1]hept-2-yl-1,1,2,2-tetrafluoroethanesulfonate, triphenylsulfonium
 2-tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl-1,1,2,2-tetrafluoroethanesulfonate,
 triphenylsulfonium N,N-bis(nonafluoro-n-butanesulfonyl)imide, triphenylsulfonium
 camphorsulfonate, 1-(4-n-butoxynaphthalen-1-yl)tetrahydrothiophenium
 trifluoromethanesulfonate, 1-(4-n-butoxynaphthalen-1-yl)tetrahydrothiophenium
 nonafluoro-n-butanesulfonate, 1-(4-n-butoxynaphthalen-1-yl)tetrahydrothiophenium
 perfluoro-n-octanesulfonate, 1-(4-n-butoxynaphthalen-1-yl)tetrahydrothiophenium
 2-bicyclo[2.2.1]heptan-2-yl-1,1,2,2-tetrafluoroethanesulfonate,
 1-(4-n-butoxynaphthalen-1-yl)tetrahydrothiophenium
 2-(tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl-1,1,2,2-tetrafluoroethanesulfonate,
 1-(4-n-butoxynaphthalen-1-yl)tetrahydrothiophenium
 N,N-bis(nonafluoro-n-butanesulfonyl)imide,
 1-(4-n-butoxynaphthalen-1-yl)tetrahydrothiophenium camphorsulfonate,
 1-(3,5-dimethyl-4-hydroxyphenyl)tetrahydrothiophenium trifluoromethanesulfonate,
 1-(3,5-dimethyl-4-hydroxyphenyl)tetrahydrothiophenium nonafluoro-n-butanesulfonate,
 1-(3,5-dimethyl-4-hydroxyphenyl)tetrahydrothiophenium perfluoro-n-octanesulfonate,
 1-(3,5-dimethyl-4-hydroxyphenyl)tetrahydrothiophenium
 2-(bicyclo[2.2.1]hept-2-yl)-1,1,2,2-tetrafluoroethanesulfonate,
 1-(3,5-dimethyl-4-hydroxyphenyl)tetrahydrothiophenium
 2-tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl-1,1,2,2-tetrafluoroethanesulfonate,
 1-(3,5-dimethyl-4-hydroxyphenyl)tetrahydrothiophenium
 N,N-bis(nonafluoro-n-butanesulfonyl)imide,
 1-(3,5-dimethyl-4-hydroxyphenyl)tetrahydrothiophenium camphorsulfonate,
 N-(trifluoromethanesulfonyloxy)succinimide,
 N-(nonafluoro-n-butanesulfonyloxy)succinimide,
 N-(perfluoro-n-octanesulfonyloxy)succinimide,
 N-(2-bicyclo[2.2.1]hept-2-yl-1,1,2,2-tetrafluoroethanesulfonyloxy)succinimide,
 N-(2-tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl-1,1,2,2-tetrafluoroethanesulfonyloxy)
 succinimide, N-(camphorsulfonyloxy)succinimide,
 N-(trifluoromethanesulfonyloxy)bicyclo[2.2.1]hept-5-ene-2,3-dicarboxyimide,
 N-(nonafluoro-n-butanesulfonyloxy)bicyclo[2.2.1]hept-5-ene-2,3-dicarboxyimide,
 N-(perfluoro-n-octanesulfonyloxy)bicyclo[2.2.1]hept-5-ene-2,3-dicarboxyimide,

N-(2-bicyclo[2.2.1]hept-2-yl-1,1,2,2-tetrafluoroethanesulfonyloxy)bicyclo[2.2.1]hept-5-ene-2,3-dicarboxyimide,
 N-(2-tetracyclo[6.2.1.1^{3,6}.0^{2,7}]dodecan-4-yl-1,1,2,2-tetrafluoroethanesulfonyloxy)
 bicyclo[2.2.1]hept-5-ene-2,3-dicarboxyimide,
 N-(camphorsulfonyloxy)bicyclo[2.2.1]hept-5-ene-2,3-dicarboxyimide and the like are preferable.

The above acid generators [B] may be used alone or in combinations of two or more.
 [0132]

The amount of the acid generator [B] in the radiation-sensitive resin composition of the present invention is usually 0.1-20 parts by mass, preferably 0.1-15 parts by mass and more preferably 0.1-10 parts by mass based on 100 parts by mass of the above resin [A]. The amount in this range ensures sufficient sensitivity and developability as a resist. If the amount of the acid generator [B] is less than 0.1 part by mass, sensitivity and developability of the resulting resist may decrease. If the amount exceeds 10 parts by mass, it may be difficult to obtain a rectangular resist pattern due to a decrease in transparency to radiation.

[0133]

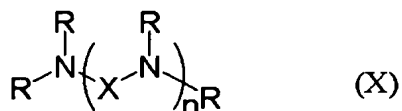
3. Additives

Various additives may be incorporated in the radiation-sensitive resin composition of the present invention. In particular, an acid diffusion controller [C] is preferably added to control diffusion of an acid generated from the acid generator [B] upon exposure in the resist film and prevents unnecessary chemical reactions in the unexposed region.

As the acid diffusion controller, a nitrogen-containing organic compound of which the basicity does not change by exposure or heating when forming a resist pattern is preferable.

[0134]

The nitrogen-containing organic compound is not particularly limited, but examples include the compound represented by the general formula (X) below, a quaternary ammonium hydroxide compound, an amide group-containing compound, an urea compound, a nitrogen-containing heterocyclic compound and the like.



(In the formula, Rs individually represent hydrogen atom, a substituted or unsubstituted, linear, branched or cyclic alkyl group, a substituted or unsubstituted aryl group, or a substituted or unsubstituted aralkyl group, X represents a divalent organic group, and n is an integer of 0-2.)

[0135]

When the substituent R in the formula (X) has a functional group, a hydroxyl group, and the like can be given. Either one functional group or two or more functional groups that are combined together may be used.

[0136]

A compound of the formula (X) in which $n=0$ is herein indicated as "nitrogen-containing compound (a)". A compound of the formula (X) in which $n=1$ or 2 is indicated as "nitrogen-containing compound (b)". Polyamino compounds and polymers having three or more nitrogen atoms are collectively referred to as "nitrogen-containing compound (c)".

[0137]

Examples of the above nitrogen-containing compounds (a) include a mono(cyclo)alkylamine such as n-hexylamine, n-heptylamine, n-octylamine, n-nonylamine, n-decylamine and cyclohexylamine; a di(cyclo)alkylamine such as di-n-butylamine, di-n-pentylamine, di-n-hexylamine, di-n-heptylamine, di-n-octylamine, di-n-nonylamine, di-n-decylamine, cyclohexylmethylamine and dicyclohexylamine; a tri(cyclo)alkylamine such as triethylamine, tri-n-propylamine, tri-n-butylamine, tri-n-pentylamine, tri-n-hexylamine, tri-n-heptylamine, tri-n-octylamine, tri-n-nonylamine, tri-n-decylamine, cyclohexyldimethylamine, dicyclohexylmethylamine and tricyclohexylamine; and an aromatic amine such as aniline, N-methylaniline, N,N-dimethylaniline, 2-methylaniline, 3-methylaniline, 4-methylaniline, 4-nitroaniline, 2,6-dimethylaniline, 2,6-diisopropyl aniline, diphenylamine, triphenylamine and naphthylamine.

[0138]

Examples of the above nitrogen-containing compound (b) include ethylenediamine, N,N,N',N'-tetramethylethylenediamine, N,N,N',N'-tetrakis(2-hydroxypropyl)ethylenediamine, tetramethylenediamine, 1,3-bis[1-(4-aminophenyl)-1-methylethyl]benzenetetramethylenediamine, hexamethylenediamine, 4,4'-diaminodiphenylmethane, 4,4'-diaminodiphenyl ether, 4,4'-diaminobenzophenone, 4,4'-diaminodiphenylamine, 2,2-bis(4-aminophenyl)propane, 2-(3-aminophenyl)-2-(4-aminophenyl)propane, 2-(4-aminophenyl)-2-(3-hydroxyphenyl)propane, 2-(4-aminophenyl)-2-(4-hydroxyphenyl)propane, 1,4-bis[1-(4-aminophenyl)-1-methylethyl]benzene, 1,3-bis[1-(4-aminophenyl)-1-methylethyl]benzene, bis(2-dimethylaminoethyl)ether, bis(2-diethylaminoethyl)ether and the like.

[0139]

Additionally, examples of the above nitrogen-containing compound (c) include polyethyleneimine, polyallylamine, a polymer of 2-dimethylaminoethylacrylamide and the like.

[0140]

Examples of the quaternary ammonium hydroxide compound used as the acid diffusion controller [C] other than the compound of the general formula (X) above include tetramethylammonium hydroxide, tetraethylammonium hydroxide, tetra-n-propylammonium hydroxide, tetra-n-butylammonium hydroxide and the like.

[0141]

Examples of the above amide group-containing compound include a N-t-butoxycarbonyl group-containing amino compound such as N-t-butoxycarbonyl di-n-octylamine, N-t-butoxycarbonyl di-n-nonylamine, N-t-butoxycarbonyl di-n-decylamine, N-t-butoxycarbonyl dicyclohexylamine, N-t-butoxycarbonyl-1-adamantylamine, N-t-butoxycarbonyl-N-methyl-1-adamantylamine, N,N-di-t-butoxycarbonyl-1-adamantylamine, N,N-di-t-butoxycarbonyl-N-methyl-1-adamantylamine, N-t-butoxycarbonyl-4,4'-diaminodiphenylmethane, N,N'-di-t-butoxycarbonylhexamethylenediamine, N,N,N',N'-tetra-t-butoxycarbonylhexamethylenediamine, N,N'-di-t-butoxycarbonyl-1,7-diaminoheptane, N,N'-di-t-butoxycarbonyl-1,8-diaminooctane, N,N'-di-t-butoxycarbonyl-1,9-diaminononane, N,N'-di-t-butoxycarbonyl-1,10-diaminodecane, N,N'-di-t-butoxycarbonyl-1,12-diaminododecane, N,N'-di-t-butoxycarbonyl-4,4'-diaminodiphenylmethane, N-t-butoxycarbonylbenzimidazole, N-t-butoxycarbonyl-2-methylbenzimidazole and N-t-butoxycarbonyl-2-phenylbenzimidazole; formamide, N-methylformamide, N,N-dimethylformamide, acetamide, N-methylacetamide, N,N-dimethylacetamide, propionamide, benzamide, pyrrolidone, N-methylpyrrolidone and the like.

[0142]

Examples of the above urea compound include urea, methylurea, 1,1-dimethylurea, 1,3-dimethylurea, 1,1,3,3-tetramethylurea, 1,3-diphenylurea, tri-n-butylthiourea and the like.

Examples of the nitrogen-containing heterocyclic compound include imidazoles such as imidazole, 4-methylimidazole, 1-benzyl-2-methylimidazole, 4-methyl-2-phenylimidazole, benzimidazole and 2-phenylbenzimidazole; pyridines such as pyridine, 2-methylpyridine, 4-methylpyridine, 2-ethylpyridine, 4-ethylpyridine, 2-phenylpyridine, 4-phenylpyridine, 2-methyl-4-phenylpyridine, nicotine, nicotinic acid, nicotinamide, quinoline, 4-hydroxyquinoline, 8-oxyquinoline and acridine; piperazines such as piperazine and 1-(2-hydroxyethyl)piperazine; pyrazine, pyrazole, pyridazine, quinoxaline, purine, pyrrolidine, piperidine, 3-piperidino-1,2-propanediol, morpholine, 4-methylmorpholine, 1,4-dimethylpiperazine, 1,4-diazabicyclo[2.2.2]octane and the like.

[0143]

Of these nitrogen-containing organic compounds, the amide group-containing compound and the nitrogen-containing heterocyclic compound are preferable. Among the amide group-containing compound, N-t-butoxycarbonyl group-containing amino compound is preferable. Among the nitrogen-containing heterocyclic compound, imidazoles are preferable. These nitrogen-containing organic compounds may be used alone or in combination of two or more.

[0144]

The amount of the above acid diffusion controller [C] to be added is usually 0.001-15 parts by mass, preferably 0.01-10 parts by mass and more preferably 0.01-5 parts by mass

based on 100 parts by mass of the above resin [A]. The addition in such amount of the above range ensures further improved storage stability of the resulting radiation-sensitive resin composition, and moreover can improve resolution as a resist and prevent the line width of the resist pattern from changing due to variation of post-exposure delay (PED) from exposure to development, whereby a composition with remarkably superior process stability can be obtained.

If the amount of the above acid diffusion controller [C] exceeds 15 parts by mass, sensitivity as a resist and developability of the exposed area tend to decrease. On the other hand, if the amount is less than 0.001 part by mass, the pattern shape or dimensional accuracy as a resist may decrease depending on the process conditions.

[0145]

Preferable proportions of the resin [A], the acid generator [B] and the acid diffusion controller [C] in the radiation-sensitive resin composition of the present invention is as follows. Based on 100 parts by mass of the above resin [A], usually 0.1-20 parts by mass of the above acid generator [B] and 0.001-15 parts by mass of the above acid diffusion controller [C], preferably 0.1-15 parts by mass of the above acid generator [B] and 0.01-10 parts by mass of the above acid diffusion controller [C] and more preferably 0.1-10 parts by mass of the above acid generator [B] and 0.01-5 parts by mass of the above acid diffusion controller [C] are added.

[0146]

Additives having an effect of improving dry etching resistance, pattern shape, adhesion to substrate and the like may be added to the radiation-sensitive resin composition of the present invention. Such additives may or may not have an acid-dissociable functional group. Examples of the additives include an adamantane derivative such as t-butyl 1-adamantanecarboxylate, t-butoxycarbonylmethyl 1-adamantanecarboxylate, α -butyrolactone 1-adamantanecarboxylate, di-t-butyl 1,3-adamantanedicarboxylate, t-butyl 1-adamantaneacetate, t-butoxycarbonylmethyl 1-adamantaneacetate, di-t-butyl 1,3-adamantanediacetate and 2,5-dimethyl-2,5-di(adamantylcarbonyloxy)hexane; a deoxycholate such as t-butyl deoxycholate, t-butoxycarbonylmethyl deoxycholate, 2-ethoxyethyl deoxycholate, 2-cyclohexyloxyethyl deoxycholate, 3-oxocyclohexyl deoxycholate, tetrahydropyranyl deoxycholate and mevalonolactone deoxycholate; a lithocholate such as t-butyl lithocholate, t-butoxycarbonylmethyl lithocholate, 2-ethoxyethyl lithocholate, 2-cyclohexyloxyethyl lithocholate, 3-oxocyclohexyl lithocholate, tetrahydropyranyl lithocholate and mevalonolactone lithocholate; an alkyl carboxylate such as dimethyl adipate, diethyl adipate, dipropyl adipate, di-n-butyl adipate and di-t-butyl adipate; and the like. Of these, t-butyl 1-adamantanecarboxylate, di-t-butyl 1,3-adamantanedicarboxylate, t-butyl 1-adamantaneacetate, 2,5-dimethyl-2,5-di(adamantylcarbonyloxy)-n-hexane, t-butyl deoxycholate, t-butoxycarbonylmethyl deoxycholate, t-butyl lithocholate, t-butoxycarbonylmethyl lithocholate and di-n-butyl adipate are preferable. These compounds may be used alone or in combination of two or more.

[0147]

The amount of the above additives to be added is usually 50 parts by mass or less, and preferably 1-30 parts by mass based on 100 parts by mass of the above resin [A]. If the amount of the above additives exceeds 50 parts by mass, heat resistance as a resist tends to decrease.

[0148]

Additionally, a surfactant capable of improving applicability, developability and the like may be added to the radiation-sensitive resin composition of the present invention. Any of an anionic surfactant, a cationic surfactant and a nonionic surfactant may be used. Of these, the nonionic surfactant is preferable. Examples include polyoxyethylene lauryl ether, polyoxyethylene stearyl ether, polyoxyethylene oleyl ether, polyoxyethylene n-octylphenyl ether, polyoxyethylene n-nonylphenyl ether, polyethylene glycol dilaurate, polyethylene glycol distearate and the like. As commercially available products, KP341 (manufactured by Shin-Etsu Chemical Co., Ltd.), Polyflow No. 75, No. 95 (manufactured by Kyoeisha Chemical Co., Ltd.), FTOP EF301, EF303, EF352 (manufactured by TOHKEM PRODUCTS CORPORATION), MEGAFAC F171, F173 (manufactured by Dainippon Ink and Chemicals, Inc.), Fluorard FC430, FC431 (manufactured by Sumitomo 3M Ltd.), Asahi Guard AG710, Surflon S-382, SC-101, SC-102, SC-103, SC-104, SC-105, SC-106 (manufactured by Asahi Glass Co., Ltd.), and the like can be given. These surfactants may be used alone or in combination of two or more.

The amount of the above surfactant to be added is usually 2 parts by mass or less and preferably 0.001-2 parts by mass based on 100 parts by mass of the total of the resin [A] and the acid generator [B].

[0149]

Further, a photosensitizer capable of improving sensitivity and the like may be added to the radiation-sensitive resin composition of the present invention. Examples include carbazoles, benzophenones, rose bengals, anthracenes, phenols and the like. These compounds may be used alone or in combination of two or more.

The amount of the above photosensitizer to be added is usually 50 parts by mass or less and preferably 1-20 parts by mass based on 100 parts by mass of the resin [A].

[0150]

Other additives to be incorporated into the radiation-sensitive resin composition of the present invention may be a halation inhibitor, an adhesion promoter, a storage stabilizer, an anti-foaming agent and the like.

[0151]

4. Solvent

The radiation-sensitive resin composition of the present invention may be one in which the resin [A], acid generator [B] and the like are dissolved in a solvent. Examples of this solvent include a linear or branched ketone such as 2-butanone, 2-pentanone, 3-methyl-2-butanone, 2-hexanone, 4-methyl-2-pentanone, 3-methyl-2-pentanone, 3,3-dimethyl-2-butanone, 2-heptanone and 2-octanone; a cyclic ketone such as

cyclopentanone, 3-methylcyclopentanone, cyclohexanone, 2-methylcyclohexanone, 2,6-dimethylcyclohexanone and isophorone; a propylene glycol monoalkyl ether acetate such as propylene glycol monomethyl ether acetate, propylene glycol monoethyl ether acetate, propylene glycol mono-n-propyl ether acetate, propylene glycol mono-i-propyl ether acetate, propylene glycol mono-n-butyl ether acetate, propylene glycol mono-i-butyl ether acetate, propylene glycol mono-sec-butyl ether acetate and propylene glycol mono-t-butyl ether acetate;

[0152]

an alkyl 2-hydroxypropionate such as methyl 2-hydroxypropionate, ethyl 2-hydroxypropionate, n-propyl 2-hydroxypropionate, i-propyl 2-hydroxypropionate, n-butyl 2-hydroxypropionate, i-butyl 2-hydroxypropionate, sec-butyl 2-hydroxypropionate and t-butyl 2-hydroxypropionate; an alkyl 3-alkoxypropionate such as methyl 3-methoxypropionate, ethyl 3-methoxypropionate, methyl 3-ethoxypropionate, and ethyl 3-ethoxypropionate;

[0153]

an alcohol such as n-propylalcohol, i-propylalcohol, n-butylalcohol, t-butylalcohol and cyclohexanol; an alkylene glycol monoalkyl ether such as ethylene glycol monomethyl ether, ethylene glycol monoethyl ether, ethylene glycol mono-n-propyl ether, ethylene glycol mono-n-butyl ether, diethylene glycol dimethyl ether, diethylene glycol diethyl ether, diethylene glycol di-n-propyl ether, diethylene glycol di-n-butyl ether, ethylene glycol monomethyl ether acetate, ethylene glycol monoethyl ether acetate, ethylene glycol mono-n-propyl ether acetate, propylene glycol monomethyl ether, propylene glycol monoethyl ether and propylene glycol mono-n-propyl ether; an aromatic solvent such as toluene and xylene; ethyl 2-hydroxy-2-methylpropionate, ethyl ethoxyacetate, ethyl hydroxyacetate, methyl 2-hydroxy-3-methylbutyrate, 3-methoxybutylacetate, 3-methyl-3-methoxybutylacetate, 3-methyl-3-methoxybutylpropionate, 3-methyl-3-methoxybutylbutyrate, ethyl acetate, n-propyl acetate, n-butyl acetate, methyl acetoacetate, ethyl acetoacetate, methyl pyruvate, ethyl pyruvate, N-methyl pyrrolidone, N,N-dimethylformamide, N,N-dimethylacetamide, benzyl ethyl ether, di-n-hexyl ether, diethylene glycol monomethyl ether, diethylene glycol monoethyl ether, caproic acid, caprylic acid, 1-octanol, 1-nonanol, benzyl alcohol, benzyl acetate, ethyl benzoate, diethyl oxalate, diethyl maleate, g-butyrolactone, ethylene carbonate, propylene carbonate and the like.

[0154]

Of these, the linear or branched ketone, the cyclic ketone, the propylene glycol monoalkyl ether acetate, the alkyl 2-hydroxypropionate, the alkyl 3-alkoxypropionate, the γ -butyrolactone and the like are preferable. These solvents may be used alone or in combination of two or more.

[0155]

5. Production of composition

The radiation-sensitive resin composition of the present invention can be generally obtained by dissolving all components in a solvent so that the total solid content is usually

3-50 % by mass and preferably 5-25 % by mass, and filtering the solution using a filter having a pore diameter of about 0.2 μm , for example.

[0156]

6. Resist pattern forming method

The radiation-sensitive resin composition of the present invention is particularly useful as a chemically-amplified resist.

In the chemically-amplified resist, an acid-dissociable group in the resin [A] dissociates by the action of an acid generated from the acid generator [B] upon exposure, thereby producing an alkali-soluble functional group represented by a carboxyl group. As a result, solubility of the exposed part of the resist in an alkaline developer increases, whereby the exposed part is dissolved in the alkaline developer and removed to obtain a positive-tone resist pattern.

[0157]

6-1. Formation of resist pattern

A resist pattern is formed from the radiation-sensitive resin composition of the present invention by applying the composition solution to a substrate such as a silicon wafer or a wafer coated with aluminum using an appropriate application method such as rotational coating, cast coating, roll coating, and spray coating to form a resist film. The resist film is then optionally pre-baked (hereinafter called "PB") and exposed to form a predetermined resist pattern. As the radiation used in this instance, ultraviolet rays, deep ultraviolet rays by KrF excimer laser (wavelength: 248 nm), ArF excimer laser (wavelength: 193 nm), F₂ excimer laser (wavelength: 157 nm) and EUV (extreme ultraviolet rays, wavelength: 13 nm, etc.), charged particle rays such as electron beams, X-rays such as synchrotron radiation, and the like can be given. Of these, deep ultraviolet rays and electron beams are preferable. The exposure conditions such as the light exposure are appropriately determined depending on the composition of the radiation-sensitive resin composition, types of additives, and the like.

It is preferable to perform post exposure bake (hereinafter called "PEB") in order to stably form a highly-accurate minute pattern. PEB ensures smooth dissociation of the acid-dissociable functional group in the resin [A]. The heating temperature for the PEB is usually in the range from 30 to 200°C and preferably from 50 to 170°C, although the heating conditions are changed depending on the content of the components in the radiation-sensitive resin composition.

To derive potential capability of the radiation-sensitive resin composition of the present invention to the maximum extent, an organic or inorganic antireflection film may be formed on a substrate used as disclosed, for example, in JP-B-H06-12452. In addition, a protective film may be provided on the resist film in order to prevent an adverse effect of basic impurities and the like that are present in the environmental atmosphere using a method described in, for example, JP-A-H05-188598.

The exposed resist film is then developed to form a prescribed resist pattern.

[0159]

6-2. Development

Although there are no specific limitations to the developer used for development, an alkaline aqueous solution prepared by dissolving at least one of alkaline compounds such as sodium hydroxide, potassium hydroxide, sodium carbonate, sodium silicate, sodium metasilicate, aqueous ammonia, ethylamine, n-propylamine, diethylamine, di-n-propylamine, triethylamine, methyldiethylamine, ethyldimethylamine, triethanolamine, tetramethylammonium hydroxide, pyrrole, piperidine, choline, 1,8-diazabicyclo-[5.4.0]-7-undecene and 1,5-diazabicyclo-[4.3.0]-5-nonene can be given. As the above alkaline compound, tetramethyl ammonium hydroxide is preferable.

The concentration of the alkaline compound in the above alkaline aqueous solution is usually 10 % by mass or less. If the concentration of the alkaline compound exceeds 10 % by mass, an unexposed part may be dissolved in the developer.

[0160]

An organic solvent may be added to the developer. As examples, linear, branched, or cyclic ketones such as acetone, methyl ethyl ketone, methyl i-butyl ketone, cyclopentanone, cyclohexanone, 3-methylcyclopentanone, and 2,6-dimethylcyclohexanone; alcohols such as methylalcohol, ethylalcohol, n-propylalcohol, i-propylalcohol, n-butylalcohol, t-butylalcohol, cyclopentanol, cyclohexanol, 1,4-hexanediol, and 1,4-hexanedimethylol; ethers such as tetrahydrofuran and dioxane; esters such as ethyl acetate, n-butyl acetate, and i-amyl acetate; aromatic hydrocarbons such as toluene and xylene; phenol, acetonitrile, acetone, and dimethylformamide can be given. These organic solvents can be used either individually or in combination of two or more.

[0161]

The amount of the above organic solvent to be added is usually 100 parts by mass or less, and preferably 0.01-20 parts by mass based on 100 parts by mass of the developer. The amount of the organic solvent exceeding 100 parts by mass may decrease developability, giving rise to a large undeveloped portion in the exposed area.

[0162]

A surfactant and the like may be appropriately added to the developer.

After development using an alkaline aqueous solution developer, the resist film is preferably washed with water and dried.

[0163]

The resist obtained from the radiation-sensitive resin composition of the present invention has sensitivity of 270 J/m^2 or less, preferably 260 J/m^2 or less, etching resistance of 0.8-1.0, and development defects of 10 or less, all determined by the methods described in Example.

[0164]

[Example]

The present invention is described below in more detail by examples. However, these examples should not be construed as limiting the present invention. It is noted that part(s) refers to part(s) by weight unless otherwise indicated.

[0165]

1. Various measuring methods

The following measuring methods and evaluation methods were used in the Examples.

(1) Weight average molecular weight (Mw) of resin [A]

The Mw was measured by gel permeation chromatography (GPC) using directly connected GPC columns (manufactured by Tosoh Corp., G2000H^{XL} x 2, G3000H^{XL} x 1, G4000H^{XL} x 1) under the following conditions. Flow rate: 1.0 ml/min., eluate: tetrahydrofuran, column temperature: 40°C, standard reference material: monodispersed polystyrene

(2) Radiation transmittance

The composition was applied onto a quartz glass plate by spin coating and pre-baked on a hot plate at 130°C for 60 seconds to form a resist film with a thickness of 0.34 µm. The radiation transmittance of the film calculated from the absorbance of light with a wavelength of 193 nm was used as a standard of transparency in a deep ultraviolet region.

[0166]

(3) Sensitivity

A silicon wafer (ARC 25, manufactured by Brewer Science) with a film having a thickness of 820 Å formed on the surface was used. A composition solution was applied to a substrate by spin coating and post-baked on a hot plate under the conditions shown in Tables to obtain resist films with a thickness of 0.34 µm. The films were exposed through a mask pattern using an ArF excimer laser exposure apparatus (manufactured by Nikon Corp., lens openings: 0.55). After performing PEB under the conditions shown in the Tables, the resist film was developed at 25°C for 60 seconds in a 2.38 % tetramethylammonium hydroxide aqueous solution, washed with water, and dried to form a positive-tone resist pattern. An optimum dose capable of forming a 0.16 µm line-and-space pattern (1L1S) with a 1:1 line width was taken as sensitivity.

[0167]

(4) Resolution

Minimum dimensions of the resist pattern resolved at the optimum dose were taken as the resolution.

(5) Dry-etching resistance

A composition was applied to a silicon wafer by spin coating and dried to form a resist film with a thickness of 0.5 µm. Then, the resist film was dry-etched using a dry-etching device (Pinnacle 8000, manufactured by PMT Co.) and CF₄ as an etching gas at a flow rate of 75 sccm and an output of 2,500 W under a gas pressure of 2.5 mTorr. A relative etching rate was evaluated based on the relative value of the etching rate of a sample to the etching rate of the film prepared using the composition of Comparative Example 1. The smaller the etching rate, the better the dry-etching resistance.

[0168]

(6) Pattern configuration

The length of the bottom (Lb) and the top (La) of a square cross-section of a

line-and-space (1L1S) pattern with a line width of 0.16 μm was measured by a scanning electron microscope. The pattern configuration was judged as "Good" when $0.85 \leq L_a/L_b \leq 1$ was satisfied and the pattern did not have a skirt-like extension, and as "Tapered" when $0.85 > L_a/L_b$. In the case of using other exposure systems, the pattern shape was evaluated by a line-and-space pattern with a line width of 0.22 μm .

[0169]

(7) Development defect

Development defect was evaluated by both a method of observing the presence or absence of the defective development using an optical microscope, and a method using a KLA defect inspection system (manufactured by KLA-TENCOR JAPAN LTD.).

The total number of defective clusters and unclusters extracted from the difference caused by superposing the pixels and a reference image was detected by observation at an array mode. The total number of defect was detected by adjusting the sensitivity of the system so that the system can detect the defects with 0.15 mm or more. In the evaluation, 12% of the total number of defects per wafer in the method of using the above-mentioned KLA defect inspection system was determined.

[0170]

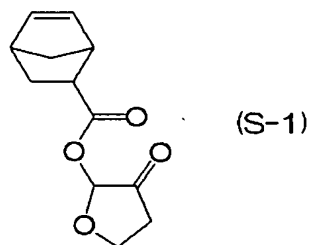
2. Production of resin [A]

(Synthesis Example 1)

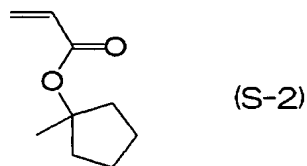
A monomer solution, prepared by dissolving 18.40 g (30 mol%) of 1-methylcyclopentyl acrylate represented by the following structural formula (S-2) and 55.09 g (40 mol%) of a compound of the following formula (S-3) in 73.48 g of 2-butanone, and further adding 7.32 g of azobis methyl isovalerate, was placed in a dropping funnel. 26.52 g (30 mol%) of the following compound (S1) and 26.52 g of 2-butanone were placed in a 500 ml three-neck flask. The mixture was purged with nitrogen for 30 minutes. After nitrogen purge, the monomer solution was added dropwise over two hours while controlling the temperature of the reaction solution at 80°C while stirring, followed by polymerization reaction for three further hours. After the polymerization, the polymer solution was cooled with water to 30°C or less. The cooled polymer solution was placed in a 1,000 ml separating funnel. After the addition of 200 g of ethyl acetate, 150 g of methanol, and 200 g of water, the mixture was stirred for five minutes and allowed to stand for one hour. Then, 250 g of the lower layer was removed. The lower layer polymer solution was replaced with a propylene glycol monomethyl ether acetate solution using an evaporator to obtain 200 g of a 39% polymer solution. The yield was 82%.

The resin obtained was a copolymer having Mw of 7,200 in which the ratio of the repeating units derived from the compound (S-1), compound (S-2), and compound (S-3) was 28.1/31.2/39.7 (mol%). This resin is referred to as "resin (A-1)".

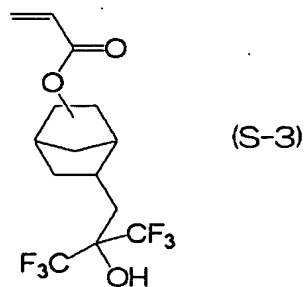
[0171]



[0172]



[0173]



[0174]

(Synthesis Example 2)

A monomer solution, prepared by dissolving 28.38 g (30 mol%) of 2-methyladamantan-2-yl acrylate and 59.48 g (40 mol%) of the above compound (S-3) in 87.87 g of 2-butanone, and further adding 7.91 g of azobis methyl isovalerate was placed in a dropping funnel. 112.13 g (30 mol%) of bicyclo[2.2.1]hept-2-ene (S-4) with the following formula and 12.13 g of 2-butanone were placed in a 500 ml three-neck flask. The mixture was purged with nitrogen for 30 minutes. After nitrogen purge, the monomer solution was added dropwise over two hours while controlling the temperature of the reaction solution at 80°C while stirring, followed by polymerization reaction for three further hours. After the polymerization, the polymer solution was cooled with water to 30°C or less, diluted with 200 g of 2-butanone, and poured into 2,000 g of methanol to obtain white precipitate. After filtration, the obtained white powder in the form of slurry was washed twice with 400 g of methanol. The white powder collected by filtration was dried at 50°C for 17 hours to obtain 75 g of a white powdery resin. The yield was 75%.

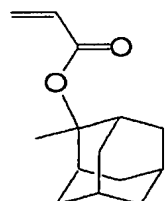
The resulting resin was a copolymer having Mw of 6,900 in which the ratio of the repeating units derived from the compound (S-4), compound (S-5), and compound (S-3) was 29.2/30.4/40.4 (mol%). This resin is referred to as "resin (A-2)".

[0175]



(S-4)

[0176]



(S-5)

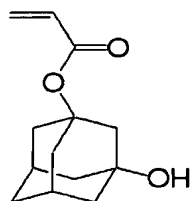
[0177]

(Synthesis Example 3)

A monomer solution, prepared by dissolving 12.54 g (20 mol%) of the above compound (S-2), 42.25 g (30 mol%) of the above compound (S-3), and 18.08 g (20 mol%) of 3-hydroxyadamantan-1-yl acrylate (S-6) in 72.88 g of 2-butanone, and further adding 7.49 g of azobis methyl isovalerate was placed in a dropping funnel. 27.12 g (30 mol%) of the above compound (S-1) and 27.12 g of 2-butanone were placed in a 500 ml three-neck flask. The mixture was purged with nitrogen for 30 minutes. After nitrogen purge, the monomer solution was added dropwise over two hours while controlling the temperature of the reaction solution at 80°C while stirring, followed by heating for three further hours. After polymerization, the polymer solution was cooled with water to 30°C or less. The cooled polymer solution was placed in a 1,000 ml separating funnel. After the addition of 200 g of ethyl acetate, 150 g of methanol, and 200 g of water, the mixture was stirred for five minutes and allowed to stand for one hour. Then, 250 g of the lower layer was removed. The lower layer polymer solution was replaced with a propylene glycol monomethyl ether acetate solution using an evaporator to obtain 200g of a 40% polymer solution. The yield was 80%.

The resin obtained was a copolymer having Mw of 64,000 in which the ratio of the repeating units derived from the compound (S-1), compound (S-2), compound (S-3), and compound (S-6) was 27.8/22.1/30.0/20.1 (mol%). This resin is referred to as "resin (A-3)".

[0178]



(S-6)

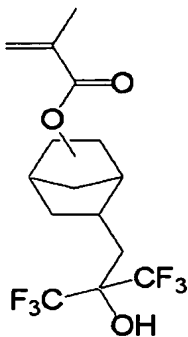
[0179]

(Synthesis Example 4)

A monomer solution, prepared by dissolving 65.27 g (55 mol%) of a compound (S-7) shown below and 34.73 g (45 mol%) of 2-methyladamantan-2-yl methacrylate (S-8) in 200 g of butanone, and further adding 3.03 g of azobis methyl isovalerate was placed in a dropping funnel. 100 g of 2-butanone was placed in a 1,000 ml three-neck flask and purged with nitrogen for 30 minutes. After nitrogen purge, the monomer solution was added dropwise at a rate of 2 ml/minute while heating the reaction solution at 80°C while stirring. The mixture was allowed to react for five hours after start of the reaction. After polymerization, the polymer solution was cooled with water to 30°C or less and poured into 2,000 g of methanol to obtain white precipitate. After filtration, the obtained white powder in the form of slurry was washed twice with 400 g of methanol. The white powder collected by filtration was dried at 50°C for 17 hours to obtain 71 g of a white powdery resin. The yield was 71%.

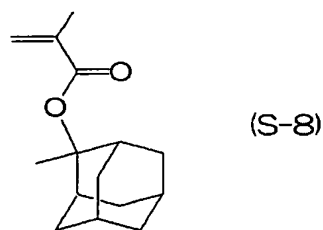
The resin obtained was a copolymer having Mw of 11,200 in which the ratio of the repeating units derived from the compound (S-7) and compound (S-8) was 57.8/42.2 (mol%). This resin is referred to as "resin (A-4)".

[0180]



(S-7)

[0181]



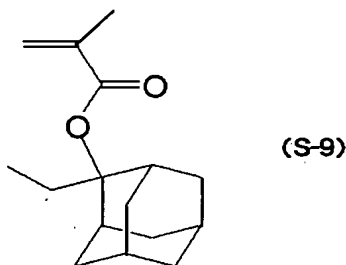
[0182]

(Synthesis Example 5)

A monomer solution, prepared by dissolving 63.94 g (55 mol%) of the compound (S-7) and 36.06 g (45 mol%) of 2-ethyladamantan-2-yl methacrylate (S-9) in 200 g of 2-butanone, and further adding 2.97 g of azobis methyl isovalerate was placed in a dropping funnel. 100 g of 2-butanone was placed in a 1,000 ml three-neck flask and purged with nitrogen for 30 minutes. After nitrogen purge, the monomer solution was added dropwise at a rate of 2 ml/minute while heating the reaction solution at 80°C while stirring. The mixture was allowed to react for five hours after start of the reaction. After polymerization, the polymer solution was cooled with water to 30°C or less and poured into 2,000 g of methanol to obtain white precipitate. After filtration, the obtained white powder in the form of slurry was washed twice with 400 g of methanol. The white powder collected by filtration was dried at 50°C for 17 hours to obtain 65 g of a white powdery resin. The yield was 65%.

The resin obtained was a copolymer having Mw of 10,900 in which the ratio of the repeating units derived from the compound (S-7) and compound (S-9) was 59.9/40.1 (mol%). This resin is referred to as "resin (A-5)".

[0183]



[0184]

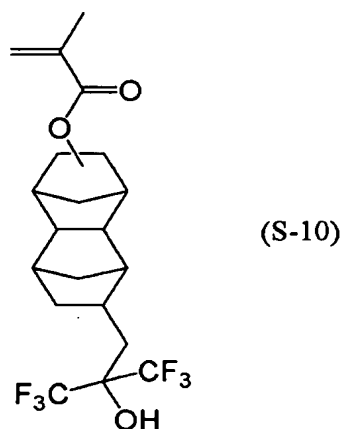
(Synthesis Example 6)

A monomer solution, prepared by dissolving 67.72 g (55 mol%) of a compound (S-10) shown below and 32.28 g (45 mol%) of the compound (S-8) in 200 g of 2-butanone, and further adding 2.66 g of azobis methyl isovalerate was placed in a dropping funnel. 100 g of 2-butanone was placed in a 1,000 ml three-neck flask and purged with nitrogen for 30 minutes. After nitrogen purge, the monomer solution was added dropwise at a rate of 2 ml/minute while heating the reaction solution at 80°C while stirring. The mixture was allowed to react for five hours after start of the reaction. After polymerization, the polymer solution

was cooled with water to 30°C or less and poured into 2,000 g of methanol to obtain white precipitate. After filtration, the obtained white powder in the form of slurry was washed twice with 400 g of methanol. The white powder collected by filtration was dried at 50°C for 17 hours to obtain 68 g of a white powdery resin. The yield was 68%.

The resin obtained was a copolymer having Mw of 11,900 in which the ratio of the repeating units derived from the compound (S-10) and compound (S-8) was 59.9/40.1 (mol%). This resin is referred to as “resin (A-6)”.

[0185]



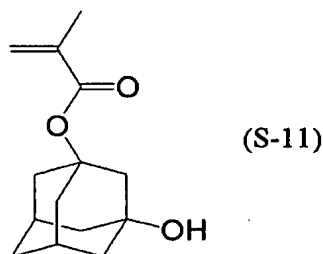
[0186]

(Synthesis Example 7)

A monomer solution, prepared by dissolving 37.69 g (25 mol%) of the above compound (S-7), 41.42 g (50 mol%) of the above compound (S-8), and 20.89 g (25 mol%) of 3-hydroxyadamantan-1-yl methacrylate (S-11) in 200 g of 2-butanone, and further adding 3.25 g of azobis methyl isovalerate was placed in a dropping funnel. 100 g of 2-butanone was placed in a 1,000 ml three-neck flask and purged with nitrogen for 30 minutes. After nitrogen purge, the monomer solution was added dropwise at a rate of 2 ml/minute while heating the reaction solution at 80°C while stirring. The mixture was allowed to react for five hours after start of the reaction. After polymerization, the polymer solution was cooled with water to 30°C or less and poured into 2,000 g of methanol to obtain white precipitate. After filtration, the obtained white powder in the form of slurry was washed twice with 400 g of methanol. The white powder collected by filtration was dried at 50°C for 17 hours to obtain 69 g of a white powdery resin. The yield was 69%.

The resin obtained was a copolymer having Mw of 9,200 in which the ratio of the repeating units derived from the compound (S-7), compound (S-8), and compound (S-11) was 29.9/45.1/25.0 (mol%). This resin is referred to as “resin (A-7)”.

[0187]



[0188]

(Synthesis Example 8)

A monomer solution, prepared by dissolving 36.78 g (25 mol%) of the above compound (S-10), 42.84 g (50 mol%) of the above compound (S-9), and 20.38 g (25 mol%) of the above compound (S-11) in 200 g of 2-butanone, and further adding 3.18 g of azobis methyl isovalerate was placed in a dropping funnel. 100 g of 2-butanone was placed in a 1,000 ml three-neck flask and purged with nitrogen for 30 minutes. After nitrogen purge, the monomer solution was added dropwise at a rate of 2 ml/minute while heating the reaction solution at 80°C while stirring. The mixture was allowed to react for five hours after start of the reaction. After polymerization, the polymer solution was cooled with water to 30°C or less and poured into 2,000 g of methanol to obtain white precipitate. After filtration, the obtained white powder in the form of slurry was washed twice with 400 g of methanol. The white powder collected by filtration was dried at 50°C for 17 hours to obtain 66 g of a white powdery resin. The yield was 66%.

The resulting resin was a copolymer having Mw of 9,400 in which the ratio of the repeating units derived from the compound (S-10), compound (S-9), and compound (S-11) was 30.2/44.2/25.4 (mol%). This resin is referred to as a "resin (A-8)".

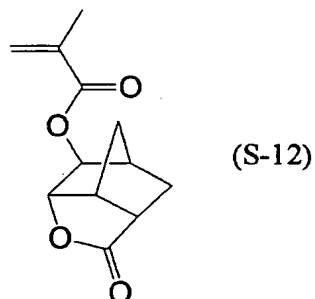
[0189]

(Synthesis Example 9)

A monomer solution, prepared by dissolving 23.06 g (16 mol%) of the above compound (S-7), 44.05 g (47 mol%) of the above compound (S-8), and 32.89 g (37 mol%) of a compound (S-12) shown below in 150 g of 2-butanone, and further adding 3.68 g of azobis methyl isovalerate was placed in a dropping funnel. 100 g of 2-butanone was placed in a 500 ml three-neck flask and purged with nitrogen for 30 minutes. After nitrogen purge, the monomer solution was added dropwise at a rate of 1.4 ml/minute while heating the reaction solution at 80°C while stirring. The mixture was allowed to react for five hours after start of the reaction. After polymerization, the polymer solution was cooled with water to 30°C or less and poured into 2,000 g of n-heptane to obtain white precipitate. After filtration, the obtained white powder in the form of slurry was washed twice with 400 g of n-heptane. The white powder collected by filtration was dried at 60°C for 17 hours to obtain 85.5 g of a white powdery resin. The yield was 85.5%.

The resin obtained was a copolymer having Mw of 12,000 in which the ratio of the repeating units derived from the compound (S-7), compound (S-8), and compound (S-12) was 15.2/44.7/50.1 (mol%). This resin is referred to as "resin (A-9)".

[0190]



[0191]

(Synthesis Example 10)

A monomer solution, prepared by dissolving 22.70 g (16 mol%) of the above compound (S-7), 36.18 g (37 mol%) of the above compound (S-9), and 41.12 g (47 mol%) of the above compound (S-12) in 150 g of 2-butanone, and further adding 3.62 g of azobis methyl isovalerate was placed in a dropping funnel. 100 g of 2-butanone was placed in a 500 ml three-neck flask and purged with nitrogen for 30 minutes. After nitrogen purge, the monomer solution was added dropwise at a rate of 1.4 ml/minute while heating the reaction solution at 80°C while stirring. The mixture was allowed to react for five hours after start of the reaction. After polymerization, the polymer solution was cooled with water to 30°C or less and poured into 2,000 g of n-heptane to obtain white precipitate. After filtration, the obtained white powder in the form of slurry was washed twice with 400 g of n-heptane. The white powder collected by filtration was dried at 60°C for 17 hours to obtain 89.9 g of a white powdery resin. The yield was 89.9%.

The resin obtained was a copolymer having Mw of 11,500 in which the ratio of the repeating units derived from the compound (S-7), compound (S-9), and compound (S-12) was 15.2/33.1/51.7 (mol%). This resin is referred to as "resin (A-10)".

[0192]

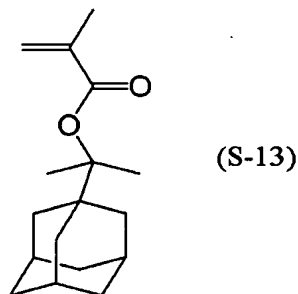
(Synthesis Example 11)

A monomer solution, prepared by dissolving 22.24 g (16 mol%) of the above compound (S-7), 37.46 g (37 mol%) of 1-adamantan-1-yl-1-methylethyl methacrylate (S-13), and 40.30 g (47 mol%) of the above compound (S-12) in 150 g of 2-butanone, and further adding 3.54 g of azobis methyl isovalerate was placed in a dropping funnel. 100 g of 2-butanone was placed in a 500 ml three-neck flask and purged with nitrogen for 30 minutes. After nitrogen purge, the monomer solution was added dropwise at a rate of 1.4 ml/minute while heating the reaction solution at 80°C while stirring. The mixture was allowed to react for five hours after start of the reaction. After polymerization, the polymer solution was cooled with water to 30°C or less and poured into 2,000 g of n-heptane to obtain white precipitate. After filtration, the obtained white powder in the form of slurry was washed twice with 400 g of n-heptane. The white powder collected by filtration was dried at 60°C for 17 hours to obtain 97.1 g of a white powdery resin. The yield was 97.1%.

The resin obtained was a copolymer having Mw of 18,000 in which the ratio of the

repeating units derived from the compound (S-7), compound (S-13), and compound (S-12) was 15.1/35.3/49.6 (mol%). This resin is referred to as “resin (A-11)”.

[0193]



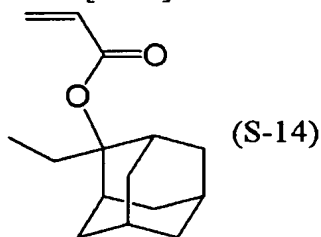
[0194]

(Synthesis Example 12)

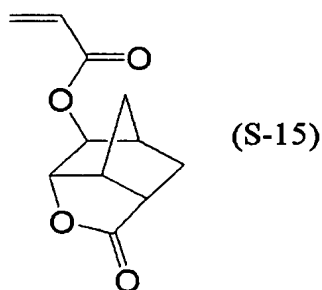
A monomer solution, prepared by dissolving 23.09 g (16mol%) of the above compound (S-3), 36.13 g(37 mol%) of 2-ethyladamantan-2-yl acrylate (S-14), and 40.78 g (47 mol%) of the above compound (S-15) in 150 g of 2-butanone, and further adding 3.84 g of azobis methyl isovalerate was placed in a dropping funnel. 100 g of 2-butanone was placed in a 500 ml three-neck flask and purged with nitrogen for 30 minutes. After nitrogen purge, the monomer solution was added dropwise at a rate of 1.4 ml/minute while heating the reaction solution at 80°C while stirring. The mixture was allowed to react for five hours after start of the reaction. After polymerization, the polymer solution was cooled with water to 30°C or less and poured into 2,000 g of n-heptane to obtain white precipitate. After filtration, the obtained white powder in the form of slurry was washed twice with 400 g of n-heptane. The white powder collected by filtration was dried at 60°C for 17 hours to obtain 90.5 g of a white powdery resin. The yield was 90.5%.

The resin was a copolymer having Mw of 10,600 in which the ratio of the repeating units derived from the compound (S-3), compound (S-14), and compound (S-15) was 15.0/34.2/50.8 (mol%). This resin is referred to as “resin (A-12)”.

[0195]



[0196]



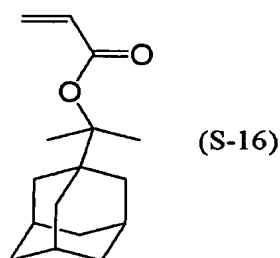
[0197]

(Synthesis Example 13)

A monomer solution, prepared by dissolving 22.60 g (16 mol%) of the above compound (S-3), 37.48 g (37 mol%) of 1-adamantan-1-yl-1-methylethyl acrylate (S-16), and 39.92 g (47 mol%) of the above compound (S-15) in 150 g of 2-butanone, and further adding 3.75 g of azobis methyl isovalerate was placed in a dropping funnel. 100 g of 2-butanone was placed in a 500 ml three-neck flask and purged with nitrogen for 30 minutes. After nitrogen purge, the monomer solution was added dropwise at a rate of 1.4 ml/minute while heating the reaction solution at 80°C while stirring. The mixture was allowed to react for five hours after start of the reaction. After polymerization, the polymer solution was cooled with water to 30°C or less and poured into 2,000 g of n-heptane to obtain white precipitate. After filtration, the obtained white powder in the form of slurry was washed twice with 400 g of n-heptane. The white powder collected by filtration was dried at 60°C for 17 hours to obtain 95.5 g of a white powdery resin. The yield was 95.5%.

The resin was a copolymer having Mw of 16,600 in which the ratio of the repeating units derived from the compound (S-3), compound (S-16), and compound (S-15) was 14.8/35.1/50.1 (mol%). This resin is referred to as "resin (A-13)".

[0198]



[0199]

(Synthesis Example 14)

A monomer solution, prepared by dissolving 23.97 g (25 mol%) of the above compound (S-10), 50.55 g (50 mol%) of the above compound (S-8), and 25.49 g (25 mol%) of the above compound (S-11) in 200 g of 2-butanone, and further adding 3.97 g of azobis methyl isovalerate was placed in a dropping funnel. 100 g of 2-butanone was placed in a 1,000 ml three-neck flask and purged with nitrogen for 30 minutes. After nitrogen purge, the monomer solution was added dropwise at a rate of 2 ml/minute while heating the reaction solution at 80°C while stirring. The mixture was allowed to react for five hours after start of the reaction.

After polymerization, the polymer solution was cooled with water to 30°C or less and poured into 2,000 g of methanol to obtain white precipitate. After filtration, the obtained white powder in the form of slurry was washed twice with 400 g of methanol. The white powder collected by filtration was dried at 50°C for 17 hours to obtain 74 g of a white powdery resin. The yield was 74%.

The resin was a copolymer having Mw of 9,800 in which the ratio of the repeating units derived from the compound (S-10), compound (S-8), and compound (S-11) was 29.2/45.2/25.6 (mol%). This resin is referred to as "resin (A-14)".

[0200]

(Synthesis Example 15)

A monomer solution, prepared by dissolving 23.97 g (25 mol%) of the above compound (S-12), 50.55 g (50 mol%) of the above compound (S-8), and 25.49 g (25 mol%) of the above compound (S-11) in 200 g of 2-butanone, and further adding 3.97 g of azobis methyl isovalerate was placed in a dropping funnel. 100 g of 2-butanone was placed in a 1,000 ml three-neck flask and purged with nitrogen for 30 minutes. After nitrogen purge, the monomer solution was added dropwise at a rate of 2 ml/minute while heating the reaction solution at 80°C while stirring. The mixture was allowed to react for five hours after start of the reaction. After polymerization, the polymer solution was cooled with water to 30°C or less and poured into 2,000 g of methanol to obtain white precipitate. After filtration, the obtained white powder in the form of slurry was washed twice with 400 g of methanol. The white powder collected by filtration was dried at 50°C for 17 hours to obtain 74 g of a white powdery resin. The yield was 74%.

The resin was a copolymer having Mw of 9,800 in which the ratio of the repeating units derived from the compound (S-12), compound (S-8), and compound (S-11) was 29.2/45.2/25.6 (mol%). This resin is referred to as "resin (a-1)".

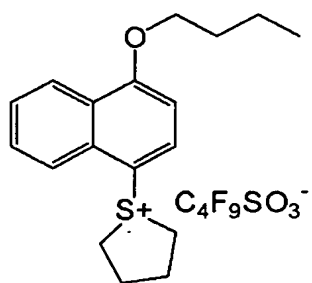
[0201]

3. Examples 1-14 and Comparative Example 1

The resins A-1 to 14 obtained above, and acid generator [B], acid diffusion controller [C], and solvent [E] indicated below were mixed at proportions shown in Tables 1 and 2 to prepare a homogeneous solution and filtered through a membrane filter with a pore diameter of 0.2 µm to obtain radiation-sensitive resin compositions. Various properties were evaluated according to the above methods. The results are shown in Tables 1 and 2.

Acid generator [B]

B-1: 1-(4-n-butoxynaphthyl)tetrahydrothiophenium nonafluoro-n-butanesulfonate



Acid diffusion controller [C]

C-1: 2-phenylbenzimidazole

Solvent [E]

E-1: propyleneglycol monomethylether acetate

	Example															
	1		2		3		4		5		6		7		8	
Resin [A]	A-1	100	A-2	100	A-3	100	A-4	100	A-5	100	A-6	100	A-7	100	A-8	100
Acid generator [B]	B-1	5	B-1	5	B-1	5	B-1	5	B-1	5	B-1	5	B-1	5	B-1	5
Acid diffusion controller [C]	C-1	0.3	C-1	0.3	C-1	0.3	C-1	0.3	C-1	0.3	C-2	0.3	C-1	0.3	C-1	0.3
Solvent [E]	E-1	600	E-1	600	E-1	600	E-1	600	E-1	600	E-1	600	E-1	600	E-1	600
Film thickness (mm)	0.34		0.34		0.34		0.34		0.34		0.34		0.34		0.34	
Substrate	ARC25		ARC25		ARC25		ARC25		ARC25		ARC25		ARC25		ARC25	
PB	130°C	90 sec	130°C	90 sec	130°C	90 sec	130°C	90 sec	130°C	90 sec	130°C	90 sec	130°C	90 sec	130°C	90 sec
PEB	120°C	90 sec	130°C	90 sec	120°C	90 sec	130°C	90 sec	110°C	90 sec	130°C	90 sec	130°C	90 sec	110°C	90 sec
Radiation transmittance (%)	71		72		72		71		70		72		70		71	
Sensitivity (J/m ²)	242		248		253		232		236		231		225		226	
Resolution (μm)	0.13		0.13		0.13		0.13		0.13		0.13		0.13		0.13	
Dry-etching resistance	0.9		0.8		0.8		0.9		1.0		0.8		1.0		0.9	
Development defects (number)	0		0		0		0		0		0		0		0	
Pattern configuration	Good		Good		Good		Good		Good		Good		Good		Good	

[0203]

Table 2

	Example										Comparative Example	
	9	10		11		12		13		14	1	
Resin [A]	A-9 100	A-10 100	A-11 100	A-12 100	A-13 100	A-14 100	a-1	100		100	100	
Acid generator [B]	B-1 5	B-1 5	B-1 5	B-1 5	B-1 5	B-1 5	B-1	5		5	5	
Acid diffusion controller [C]	C-1 0.3	C-1 0.3	C-1 0.3	C-1 0.3	C-1 0.3	C-1 0.3	C-1	0.3		0.3	0.3	
Solvent [E]	E-1 600	E-1 600	E-1 600	E-1 600	E-1 600	E-1 600	E-1	600		600	600	
Film thickness (mm)	0.34	0.34		0.34		0.34		0.34		0.34	0.34	
Substrate	ARC25	ARC25		ARC25		ARC25		ARC25		ARC25	ARC25	
PB	130°C 90 sec	130°C 90 sec	130°C 90 sec	130°C 90 sec	130°C 90 sec	130°C 90 sec	130°C	90 sec		130°C 90 sec	130°C 90 sec	
PEB	120°C 90 sec	120°C 90 sec	100°C 90 sec	100°C 90 sec	100°C 90 sec	100°C 90 sec	100°C	90 sec		100°C 90 sec	130°C 90 sec	
Radiation transmittance (%)	72	71		72		71		72		71	70	
Sensitivity (J/m ²)	290	310		272		240		264		226	274	
Resolution (μm)	0.13	0.13		0.13		0.13		0.13		0.13	0.13	
Dry-etching resistance	0.9	0.9		0.9		0.9		1.0		1.0	1.0	
Development defects (number)	0	0		0		0		0		0	526	
Pattern configuration	Good	Good		Good		Good		Good		Good	Good	

[0204]

[Effect of the Invention]

The radiation-sensitive resin composition of the present invention is useful as a chemically-amplified resist sensitive to active rays, particularly deep ultraviolet rays represented, for example, by a KrF excimer laser (wavelength 248 nm) or an ArF excimer laser (wavelength: 193 nm). In addition, the radiation-sensitive resin composition of the present invention exhibits high resolution and excels in sensitivity, pattern shape, and etching resistance due to the high radiation transmittance. Furthermore, the radiation-sensitive resin composition of the present invention produces development defects only to a minimal extent due to small fluctuation of patterns after etching and excellent solubility, particularly, in developing solutions. The composition also exhibits excellent adhesion with substrates and produces a superior skirt configuration. Therefore, the radiation-sensitive resin composition of the present invention is very suitable for manufacturing integrated circuit elements which are demanded to become further miniaturized in the future.

[DOCUMENT NAME]

ABSTRACT

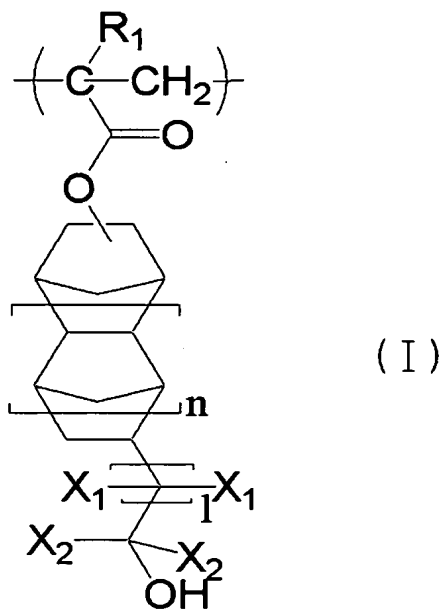
[ABSTRACT]

[PROBLEM TO BE SOLVED]

A radiation-sensitive resin composition which is useful as a chemically amplified resist in microfabrication with various radiations including deep ultraviolet rays such as KrF excimer laser and ArF excimer laser is provided.

[MEANS FOR SOLUTION]

The composition comprises [A] a resin comprising repeating units represented by the general formula(I-1), [B] a radiation-sensitive acid generator (such as 1-(4-n-butoxynaphthyl)tetrahydrothiophenium nonafluoro-n-butanesulfonate). Further, [C] an acid diffusion controller (such as phenylbenzimidazole) may be contained.



[SELECTED DRAWING] None